A Comprehensive Review of Plasma Gasification as a Promising Solution for Waste-to-Hydrogen Conversion

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Abstract

The growth of municipal solid waste (MSW) generation, coupled with diminishing natural resources, has created an urgent need for sustainable solutions to minimize environmental impact and reduce reliance on raw material extraction. Traditional waste disposal methods, including landfilling and conventional gasification, present significant limitations in terms of operational efficiency, environmental pollution, and carbon emissions. A comprehensive evaluation of waste-to-hydrogen pathways, beginning with an overview of MSW generation and traditional disposal methods, focusing on landfills, landfill gas recovery, and conventional decomposition techniques such as gasification, which produce synthesis gas and other derivatives. Then, various hydrogen production methods, categorizing them as grey, blue, green, etc, are discussed in detail based on their carbon footprint and production processes. Plasma pyrolysis/gasification technology identifies as a promising approach for low-carbon and carbon-neutral hydrogen production, offering higher efficiency and lower environmental impact compared to conventional methods. A detailed analysis of plasma technology follows, covering different plasma sources (DC, AC, RF-ICP, microwave, and hybrid) and their applications in waste-to-hydrogen processing. The article further discusses the shortcomings in adapting present plasma technology for waste processing, along with the technical, economic, and policy considerations essential for the commercialization and large-scale adoption of plasma-based waste management systems. A sustainable waste circulation model is proposed, integrating waste generation, conversion into energy, and resource recovery in a closed-loop system for a holistic net zero concept. By addressing the limitations of conventional methods and recommending plasma technology as a scalable and eco-friendly alternative, this article contributes to advancing waste management solutions and clean energy production.

Keywords: MSW; Landfill gas; Synthesis gas; Thermal plasma; Plasma gasification; Hydrogen

1. Introduction

The human living standards are the principal catalyst for the rapid progression of urbanization and industrialization. As per the United Nations (UN) urbanization status report, the number of people living in urban areas surpassed the number living in rural ones in the year 2007 [1]. These activities have resulted in an increase in raw material consumption, responsible for the loss of land use biodiversity and greenhouse gas (GHGs) emissions. The global material extraction of approximately 31 million tons in 1970 increased to 95 million tons in the year 2020, and studies estimate a growth of 60% by 2060 in the absence of any major policy changes. The higher extraction of material and processing is the reason for the 55% of the total GHGs, loss of land use biodiversity up to 90%, and studies are showing an increase in loss of 45% more by 2060 [2]. Overconsumption of material and uncontrolled management led to the humongous volume of waste generation. The most common waste generated on a daily basis is municipal solid waste (MSW), and its presence can be seen in our surroundings in the form of small open dumpsites to gigantic mountains. The current world population is 8 billion, and studies show that 3 billion people still dump their waste in an uncontrolled waste disposal facility, and 2 billion people still don’t have access to waste collection facilities. The global MSW generation is more than 2 billion tons, and this volume is expected to increase up to 3.8 billion tons by 2050 without taking any further actions [2,3]. Plastic waste, one of the known fractions of MSW, is a major cause of marine litter. Approximately 8 million tons of plastic waste are already

dumped in marine environments, challenging the survival of various species. Plastic is made from crude oil, which, after single or multiple uses, loses its properties and needs some chemical/physical treatment for reuse.

Today, human activities have exploited the natural resources for their comfort, and mismanagement has resulted in millions of tons of waste in our surroundings. Figure 2 illustrates the proper management of wasted plastic (4-R principle to protect natural resources) and how we are now handling it, which eventually results in a new food chain that threatens the life of the issue originator itself. Similarly, human activities have worsened the situation over the years, and now MSW management is a global burning topic in research as well as in our day-to-day discussions. An analysis of the trend in the publication and number of countries in collaboration is working in the field of solid waste management, waste to energy, and plasma gasification is done using Visually scientific landscape (VOS) viewer software. The data regarding publication in the given topic is collected through Dimension AI tools. The minimum number of papers published in a country is taken as one, and the trend in the last year is studied. Here is a summary of the analysis, where the significant growth in the plasma gasification research can be observed. Solid waste management and waste-to-energy is a vast field which includes all types of waste generated, which is managed and converted to energy or other usable products/byproducts. Figure 1 (a), (b) and (c) give information about the countries doing research on solid waste management, waste-to-energy and plasma gasification, respectively, separately or in collaboration. Figure 1(d) gives a brief idea about how the trend is in the number of publications in the last ten years for the respective topics. The publication numbers show the demand for the topic, for waste management and energy generation, where a linear increment can be seen over the years. Whereas for plasma gasification, a sharp rise (approximately four times in the last ten years) in publications can be seen, which shows the necessity of the topic.

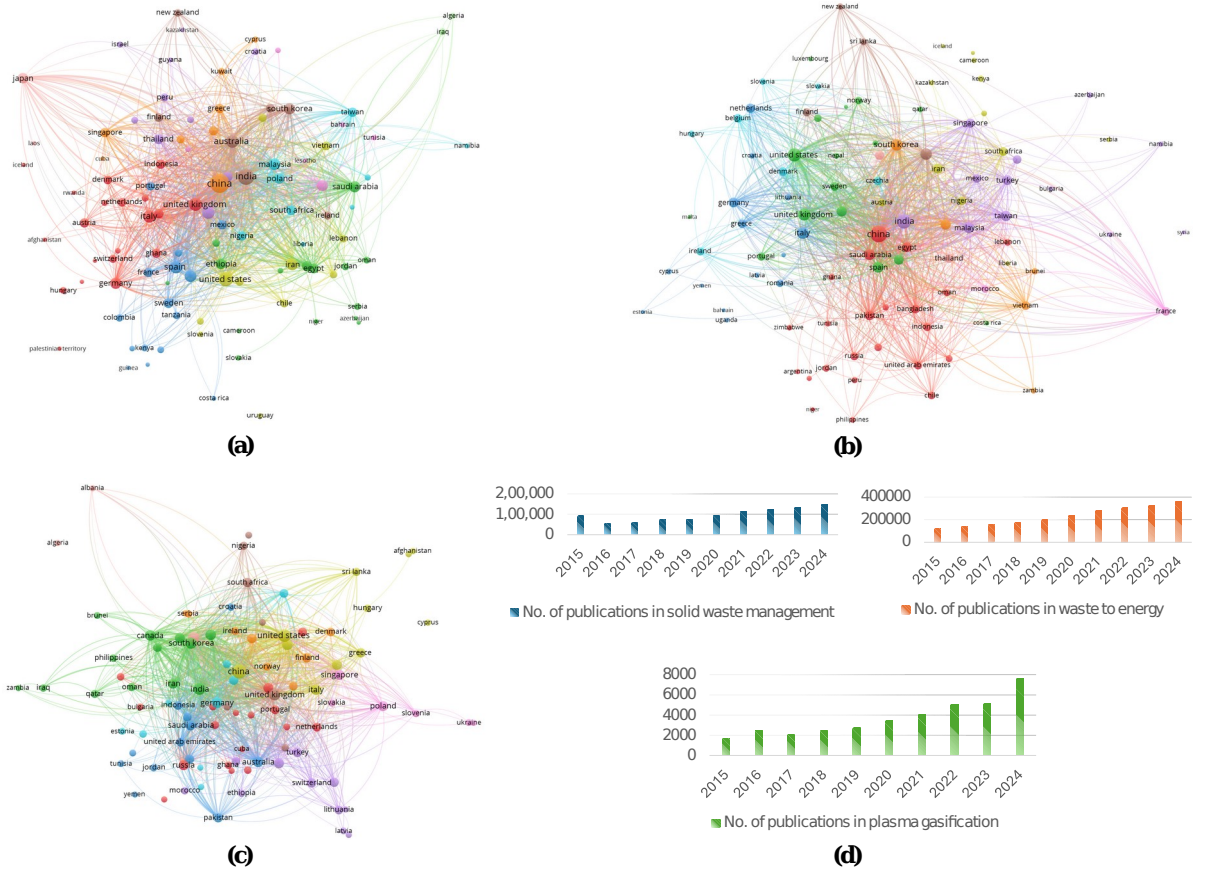

Figure 1. Network visualization for research article publications in (a) solid waste management, (b) waste to energy, (c) plasma gasification, and (d) trends in the numbers of publications over the last ten years for the respective topics [4].

It is high time to give a thought to controlling waste generation and resource waste through some sustainable practices. The generated waste can be converted into useful products and byproducts, which might limit the extraction up to significant limits of the demands. This review article discusses the same focus on different aspects of MSW generation to their management using conventional technology and how plasma technology (an advanced waste management technology) can be a better solution, overcoming the challenges of the conventional one. This article includes several sections, and it starts with MSW generation, and the basic and oldest methods of waste disposal known as landfills. Where it includes a detail of landfill gas and other conventional methods of decomposition, such as gasification and others, for synthesis gas and then other related products and by-products. Section 2 includes an in-depth review of hydrogen, types of hydrogen, and their production methods, with pros and cons. The review on hydrogen suggests plasma technology as one of

the upcoming solutions for low-carbon and carbon-neutral hydrogen production. Section 3 gives a detailed review of plasma technology and different plasma sources for various waste processing. Section 4 is a discussion of the entire review done in the article, where the technical, economic, and policy scope for the commercialization success of technology is discussed. A sustainable solution is proposed that includes the entire chain of waste circulation, i.e., waste generation to conversion into energy and other useful products without producing any secondary waste. This review article will be the first of its kind where thermal plasma technology is discussed in detail for hydrogen production and things to do for successful commercialization.

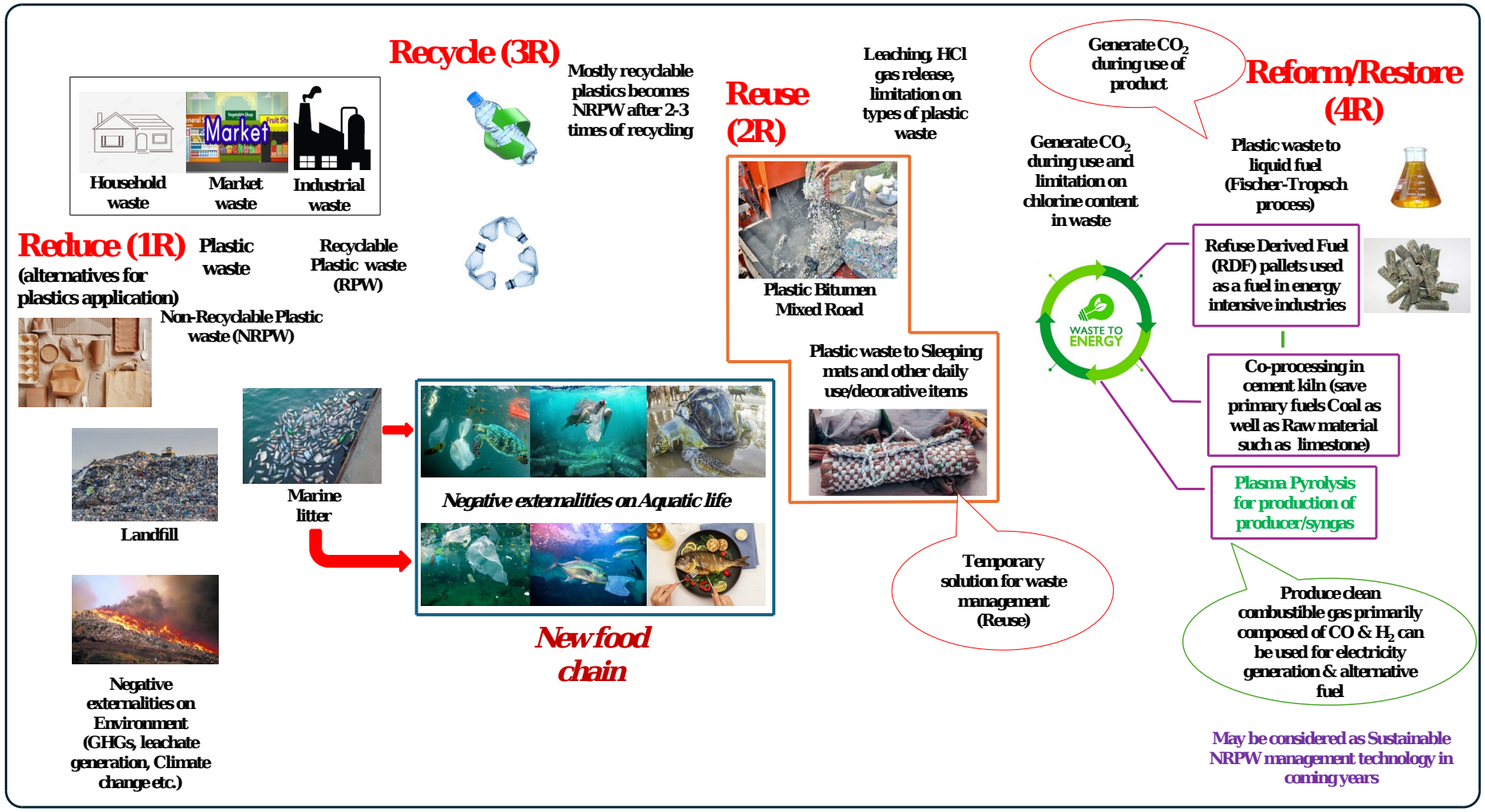
Recycle (3R)
Mostly recyclable plastics becomes NRPW after 2-3 times of recycling
Reuse (2R)
Leaching, HCl gas release, limitation on types of plastic waste
Generate CO_2 during use of product
Reform/Restore (4R)
Household waste
Market waste
Industrial waste
Reduce (1R)
(alternatives for plastics application)
Plastic waste
Recyclable Plastic waste (RPW)
Non-Recyclable Plastic waste (NRPW)
Plastic Bitumen Mixed Road
Generate CO_2 during use and limitation on chlorine content in waste
Plastic waste to liquid fuel (Fischer-Tropsch process)
WASTE TO ENERGY
Refuse Derived Fuel (RDF) pallets used as a fuel in energy intensive industries
Co-processing in cement kiln (save primary fuels Coal as well as Raw material such as limestone)
Plasma Pyrolysis for production of producer/syngas
Plastic waste to Sleeping mats and other daily use/decorative items
Temporary solution for waste management (Reuse)
Produce clean combustible gas primarily composed of CO & H_2 can be used for electricity generation & alternative fuel
Landfill
Marine litter
Negative externalities on Aquatic life
New food chain
Negative externalities on Environment (GHGs, leachate generation, Climate change etc.)
May be considered as Sustainable NRPW management technology in coming years

Figure 2. Plastic waste management using the 4R principle, avoiding the new food chain due to mismanagement.

1.1. Municipal solid waste

The waste generated from residences, retailers, small businesses, and public service providers are commonly known as municipal solid waste (MSW). It does not include waste generation sources such as construction & demolition activity, industry, agriculture, healthcare, etc. MSW includes multiple materials such as food, packaging, broken furniture, textiles, plastics, etc. The composition of MSW can't be the same for all the places, as it depends on the consumption behaviour, standard of living, population density, etc. The global average MSW composition, according to the UN report is given in Figure 3 [5]. The data collection for the first MSW generation begins with 2.4 kg per day in the region of North America in 6500 BC [6]. Today we are generating approximately 2.1 billion tons of waste worldwide annually, and the Global Waste Management Outlook 2024 predicts an increment of 56 % (3.8 billion tons) by 2050 without taking any preventive measures. The forecasting studies are considering responsible factors such as growth in gross domestic product (GDP) and population for the addition in waste generation. The GDP growth is responsible for approximately a billion tons of waste, while population growth is half a billion tons increase in waste generation [5].

The generation of MSW with such a massive quantity needs proper management. However, we are lagging far behind and disposing of lots of waste in an uncontrolled manner. The uncontrolled disposal of waste is defined as collected waste burned or dumped in an open or uncollected waste burner or open dumped. As per the United Nations Environment Programme (UNEP) report 38 % of generated waste gets disposed in an uncontrolled manner and 62 % of controlled waste management practice includes landfill (19 %), waste to energy (13 %) and recycling (30 %). The study is also predicting a rise in uncontrolled waste disposal by 2050 up to 50% if not taking any preventives [2,5]. Uncontrolled waste management degrades land, groundwater and air quality, leading to health risks to humans, animals and the ecosystem. The daily

accumulation of waste materials is causing a scarcity of further land to be used for landfills. These landfills are generally without any scientific treatment or segregation in developing countries, including India. As a result, these landfill sites have become a breeding ground for contamination by flies, rodents, and mosquitoes, causing diseases like dermatological, respiratory, genetic, and several other kinds of infectious diseases [7]. In addition to contamination, these locations are now also a significant source of greenhouse gases (GHGs) emission through the release of gases generated in landfills, also known as landfill gas (LFG). The LFG generation process and solutions required are discussed in detail in the subsequent section.

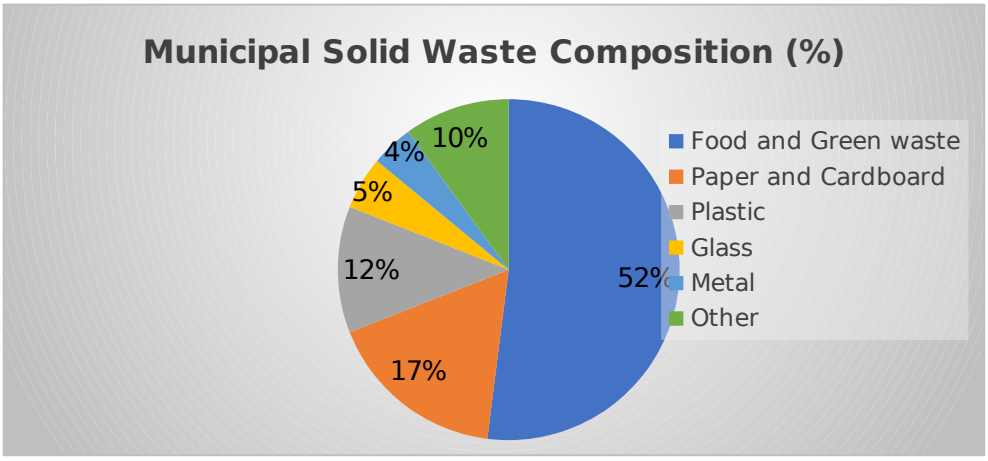

Figure 3. Global average municipal solid waste composition [2].

1.2. Landfill Gas

LandfillsFigure 3 are widely employed for solid waste disposal worldwide, particularly in developing nations such as China, Mexico, and India, where they serve as the primary method due to advantages like cost-effectiveness and minimal technological requirements [8]. While landfilling remains a common practice, it should be considered the least preferred option for solid waste management owing to its high greenhouse gas (GHG) emissions compared to alternative waste management technologies [9]. Municipal waste treatment plants are often referred to as "bioreactors" due to the natural transformation of organic matter within a confined space, leading to the formation of biogas/LFG [10,11]. LFG primarily consists of CH_4 (50–55%) and CO_2 (40–

45%) with minor amounts of other gases like nitrogen (N_2), produced during the anaerobic degradation of solid biodegradable organic waste dumped in landfills [12]. Once MSW is deposited in a landfill, the organic components begin to undergo biochemical reactions. Near the landfill surface, where atmospheric air is present, natural organic compounds are oxidized aerobically, producing carbon dioxide (CO_2) and water vapour (H_2O) in a process analogous to combustion [13,14]. Methane gas generation from landfills is the second most common GHG with a global warming potential of 28 - 34 times greater than that of CO_2 [15,16]. LFG is not a significant concern for developed countries like the USA and European countries as they are capable of collecting these harmful gases and converting them into an energy source, producing electricity [17,18]. Whereas, for developing countries, it will become a critical issue in the coming years as forecasted by studies, where developing countries like India are predicted to be major contributors of GHGs emissions through LFG with a 20 Mg CO_2 equivalent methane emission rate (current rate is 16 Mg CO_2 equivalent) [19].

Energy recovery from LFG presents a viable solution for generating energy from renewable sources [20]. The utilization of CH_4 present in LFG through combined heat and power (CHP) systems offers an environmentally and energetically efficient approach [21,22]. With the global increase in waste generation projected to accelerate in the coming years, addressing landfill reclamation has become imperative [23]. The energy recovery method involves the collection and utilization of LFG produced in landfills. Classified as a renewable energy source, LFG is typically purified and combusted in cogeneration units to convert its energy into electricity and heat. Although these gases contain a significant proportion of methane, its energy value is reduced by the presence of carbon dioxide. Extraction from landfills requires specialized shafts for gas collection, which can then be processed for use in generating electricity and heat. LFG with methane content exceeding 40% can also serve as a fuel for gas turbines, replacing conventional fuels like natural gas or propane-butane [24]. Landfill degassing can be

achieved using two methods, such as passive degassing and active degassing. However, these methods typically achieve only about 50% of the landfill's biogas production potential. Regardless of the method, LFG must be treated to remove moisture and solid particles, then compressed for further use. After initial purification, the processed biogas is either utilized in CHP systems for electricity and heat generation or burned directly in boilers.

However, the major problems associated with the method are the presence of impurities in LFG, such as corrosive substances, which can damage mechanical components in gas engines used for electricity production [20]. Looking at these problems, advanced technologies are required with more efficient output that may overcome the technical limitations. The application of advanced technology like plasma pyrolysis/gasification, capable of converting MSW into a high calorific value product (synthesis gas/hydrogen) with negligible impurities and reusable byproducts (ash/vitrified slag) with minimal environmental disturbance should be preferred [25]. This is discussed in detail in the subsequent section.

1.3. Synthesis Gas

The gas compositions rich in CO and H are important gases that have various applications in industries since the early 1900s. The production started using raw material as coke, where some of the coke is burned in the presence of air to heat the bed to 1000 °C, with a supply of blasts of air and steam. The exothermic reaction of coke with air heats the bed, and the endothermic reaction of coke with steam converts it into CO and H_2, the concentration of CO_2 at 1000°C is found to be insignificant, as given in Eqs. (1) and (2). Further, Eqs. (3) is an exothermic reaction, also known as a water gas shift (WGS) reaction that converts CO into hydrogen, reacting with steam. Since this mixture of gases has found various applications in the synthesis of chemicals such as ammonia, methanol, hydrocarbon fuels, etc, thus termed as synthesis gas (in short, syngas) [26].

$$C + H_2O \rightarrow CO + H_2 \qquad \Delta H_{298°C} = +31 \text{ kcal} \quad (1)$$

$$C + 2H_2O \rightarrow CO_2 + 2H_2 \qquad \Delta H_{298°C} = +21 \text{ kcal} \quad (2)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad \Delta H_{298°C} = -10 \text{ kcal} \quad (3)$$

The application of synthesis gas started with the extraction/separation of hydrogen from the mixture and used for the catalytic synthesis of various chemicals like ammonia (NH_3) and methanol (CHOH), as shown in Eqs. (4) and (5). The catalyst used in the catalytic synthesis of methanol is a mixture of zinc oxide and chromia [27].

$$N_2 + 3H_2 \rightarrow 2NH_3 \quad (4)$$

$$2H_2 + CO \rightarrow CH_3OH \quad (5)$$

Steam reforming, a highly endothermic process, was introduced in the 1920s, where coke is replaced with natural gas, and production starts with the conversion of natural gas into synthesis gas [28,29]. The advantages of natural gas over coke are a higher calorific value and being comparatively cleaner than coke, which leads to an increase in hydrogen concentration and overall efficiency of the process, with a significant reduction in CO_2 emissions. The steam reforming process takes place at a higher steam to methane ratio in the range of 2–4, responsible for the production of 3 times more hydrogen production than the coke-based production, as shown in Eq. (6). Later in the 1950s, some advancement was made in the process known as autothermal reforming. It is a combination of partial oxidation and steam reforming that takes place in primary and secondary chambers, respectively. The partial oxidation includes the supply of oxygen to natural gas for heat generation to maintain a temperature of more than 800 °C for steam reforming in a secondary chamber [26,30].

$$CH_4 + H_2O \rightarrow CO + 3H_2 \qquad \Delta H_{298°C} = +49 \text{ kcal} \quad (6)$$

Syngas is traditionally produced with raw materials such as natural gas, coal, coke, oil/residue and petroleum. However, studies show biomass as a more potential and sustainable candidate than fossil fuel for the benefits of producing carbon-negative products and by-products [31,32]. Biomass is a biodegradable and renewable source of energy that includes material of biological origin, such as food, forest residue, agricultural byproducts, living microorganisms, algae, animal waste, etc. The syngas generated from biomass is known as bio-syngas. Lignocellulosic material such as sugarcane, corn straw, rice straw, etc., are commonly used for bio-syngas due to a higher rate of thermal conversion into H_2 and CO [33–35]. Biomasses are mainly converted into bio-syngas through thermochemical and biochemical processes, which transform solid waste into vapours (condensable and non-condensable) and residue (ash/char). The condensable materials are known as tar (aromatic compounds such as toluene, naphthalene, benzene, etc.) and bio-oil (complex hydrocarbon compounds with a higher number of carbons than tar). The non-condensable gases are composed of long-chain heavy hydrocarbon gases (C_2-C_4); thus, they need further transformation into lighter hydrocarbon gases. Reformers have been used to crack the heavy compounds into raw syngas, which mainly consists of H_2 and CO, and this is the main technical challenge involved, including the energy-intensive process [32,36]. Reforming processes involved in the conversion of a complex compound to bio-syngas are dry reforming, steam reforming, autothermal reforming, partial oxidation, catalytic partial oxidation, membrane reforming, etc. Gasification and pyrolysis are two major thermochemical processes used for the production of bio-syngas. Higher temperature treatment is recommended for optimum bio-syngas yield. Gasification is generally taking place at higher temperatures (>1000 °C) where fuel (biomass) is fed into a reactor with a gasifying agent (air-air gasification, O_2-oxygen gasification, or H_2O-steam gasification) and undergoes thermochemical conversion.

The presence of higher temperature leads to a lower yield of solid by-products, products, resulting in a higher rate of conversion of biomass to bio-syngas than the pyrolysis process [32,37,38]. The major challenge of these thermochemical processes is tar formation in the form of aerosol. It is very difficult to separate, as it leaves the chamber with the product gases, sticks to the downstream component during condensation and leads to fouling and blockage of downstream components. The complexity of tar presents a challenging task for further breakdown into smaller molecules, and deactivation of the catalysts reduces the efficiency of conversion processes such as methanation, Fischer-Tropsch synthesis (F-T), etc. [39,40].

Syngas generation from various raw material streams has applications in the production of a wide range of chemical products. The most common route is synthesis gas to methanol, and methanol gets converted into various chemicals such as formaldehyde, methyl acetate, acetic acid, ethylene propylene, dimethyl ether (DME), etc. [31,41,42]. Another famous route is Fischer-Tropsch (FT) synthesis, which has been used widely for the production of hydrocarbon fuels, lubricating oils and oxygenated hydrocarbons such as wax, diesel, kerosene, gasoline, naphtha, etc. The FT method includes a heterogeneous catalytic reaction where metal catalysts that are prone to CO dissociation, such as cobalt, iron, ruthenium, etc., are selected [42,43]. These reactions take place in two parts; the initiation with the breakdown of the main molecular structure leads to the generation of their monomers, and then polymerisation of them produces synthetic oil with larger chemical structures [44,45]. FT synthesis is considered a sustainable method for the production of high-performance synthetic liquid fuel with the benefit of improvement in energy efficiency of internal and external combustion engines and a low carbon footprint. Syngas can also be directly used for power generation through gas turbine engines, internal combustion engines, and fuel cells; however, impurities and lower calorific value are still issues that limit its direct application. The production of syngas from various raw materials with different applications is detailed in Figure 4.

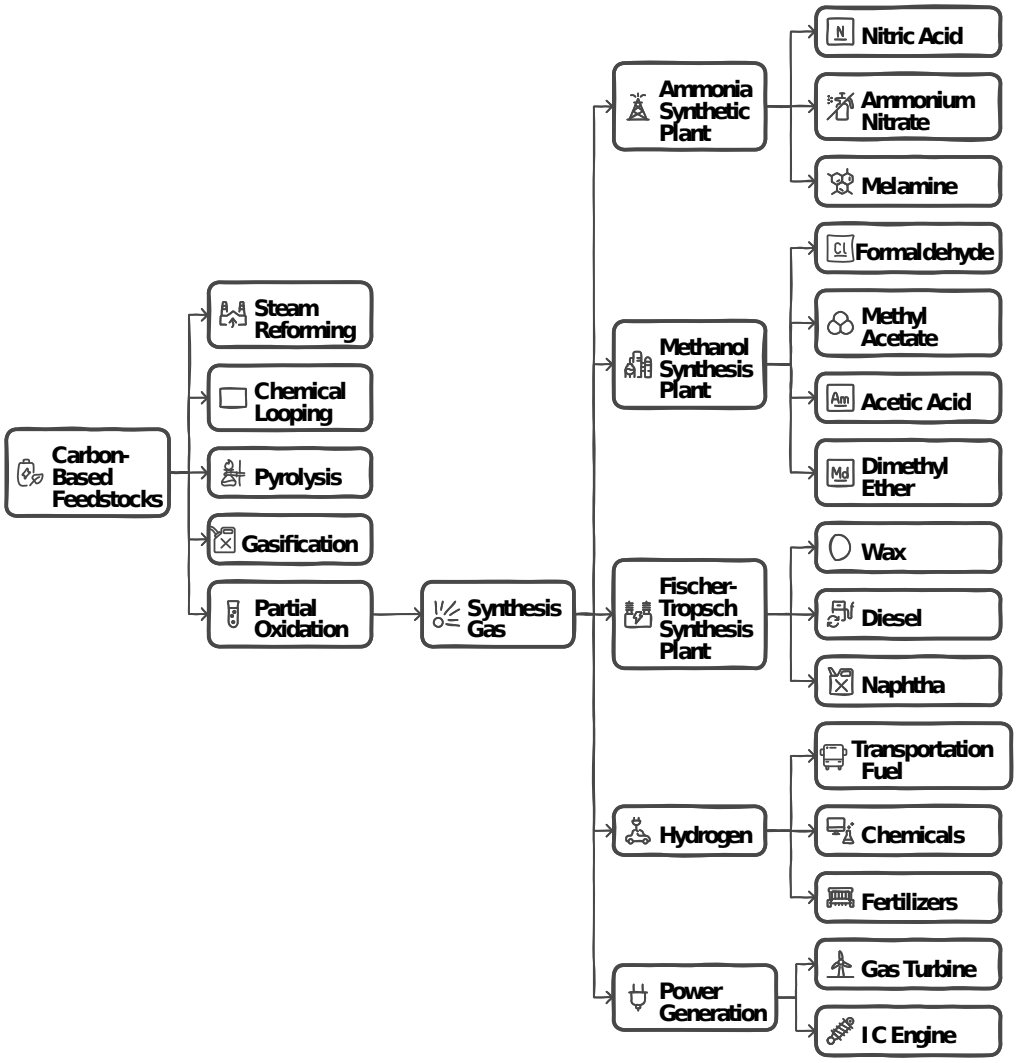

Figure 4. Synthesis gas production methods, products, and various applications *[46,47]*.

Out of all the products and byproducts produced from synthesis gas, hydrogen is one of the cleanest and most carbon-neutral fuels. In the coming years, reducing the carbon footprint and restoring climatic conditions will be more environmentally demanding, which will be discussed in detail in the following sections.

2. Hydrogen

It is the most abundant element in the universe and is primarily present on Earth in water and organic compounds. It is the lightest and simplest element, consisting of a single electron and proton [48,49]. Hydrogen is a colourless, odourless, and flammable gas, first identified through the reaction between sulfuric acid and iron [50]. It is non-toxic and significantly lighter than air, allowing it to disperse rapidly in the event

of a leak, which enhances its safety compared to other liquid fuels [51]. Its energy content is remarkably high, with a higher heating value (HHV) of 141.8 MJ/kg and a lower heating value (LHV) of 120 MJ/kg at 298 K, surpassing most conventional fuels [52,53]. However, liquid hydrogen's energy density by volume is considerably lower (8 MJ/L) than that of hydrocarbons, such as gasoline (32 MJ/L) [54]. While hydrogen offers superior energy density by weight, its lower volumetric energy density necessitates larger storage tanks for practical applications [55]. Hydrogen’s flammability and relatively low ignition temperature present challenges related to its safe handling and usage. Despite these risks, its potential to integrate seamlessly into existing energy and transportation infrastructures, coupled with its zero-carbon emissions at the point of use, positions hydrogen as a promising solution for reducing air pollution and transitioning to a cleaner energy of future [53,56]. In recent years, hydrogen demand has increased, and massive installation is projected globally to fuel the transportation sector through hydrogen fuel cells and hybrid vehicles [57].

Hydrogen could function as a crucial fuel in the current scenario of rising global temperatures and rampant carbon emissions. The benefits of hydrogen include being an excellent energy carrier, a clean fuel with zero GHG emissions, higher conversion energy efficiency, zero carbon emission-based production through water electrolysis, a feedstock for synthesis fuel and ammonia production, and faster refuelling time [57,58]. It can be produced utilizing both renewable and non-renewable sources [59]. Non-renewable sources, including coal, coke, nuclear energy, and natural gas, generate hydrogen through methods such as steam reforming, hydrocarbon reforming, partial oxidation, autothermal reforming, gasification, and pyrolysis [60–62]. Renewable sources, including biomass, solar, wind, hydro, tidal, and geothermal energy, are prospective alternatives to fossil fuels with a minimal carbon footprint [57,63–66]. These sources provide alternatives for utilizing energy in hydrogen generation instead of direct usage. The advantages of this approach include achieving net zero carbon emissions, as the utilization

of hydrogen for power generation and other operations primarily produces water as the principal byproduct. Renewable sources can generate hydrogen through methods like electrolysis, photoelectrochemical processes, photocatalysis, and thermochemical techniques. Biomass can also be transformed into hydrogen through biological processes like bio-photolysis, dark fermentation, and photo-fermentation, as well as thermochemical methods such as gasification, pyrolysis, partial oxidation, and liquefaction. Electrolysis is a renewable method for hydrogen production that can be conducted using several techniques, including alkaline, solid oxide, proton exchange membrane electrolysis, etc. [35,57,59,60].

Although hydrogen is a colourless gas, various production methods necessitate the use of colour coding for enhanced comprehension. Hydrogen is categorized into over ten distinct colour codes, including white, black, brown, orange, grey, blue, pink, red, purple, yellow, turquoise, and green, as detailed in Figure 5. Naturally occurring hydrogen, called white hydrogen, is produced by ongoing geological reactions in hard rock located in subterranean deposits. This type of hydrogen is lightweight and poses low carbon intensity (0.4 kg CO_2/kg H_2). This form of hydrogen is naturally abundant, yet extraction is challenging. The injection of a mix of water, sand, and chemicals into geological strata under high pressure facilitates the extraction of gas from the rocks, known as the fracking process. The adverse externalities of the extraction process include groundwater contamination, methane emissions, and habitat loss. Stringent safety measures are implemented during extraction due to the flammability of hydrogen gas. Aside from white hydrogen, all other forms of hydrogen are artificially manufactured through diverse ways utilizing both renewable and non-renewable resources. Gasification transforms fossil fuels, such as coal, into hydrogen [67]. Coal gasification utilizing black coal (bituminous coal: C – 45-86%) and brown coal (lignite: C – 25-35%) is referred to as black and brown hydrogen, respectively, and produces substantial CO_2 emissions (19-30 kg CO_2/kg H_2) [68]. The gasification method utilizing plastic for

hydrogen synthesis, referred as orange hydrogen, is in the nascent stages of development, having the benefit of less energy input than coal gasification [69]. Natural gas is transformed into hydrogen using the steam methane reforming method, classified as grey hydrogen, which emits greenhouse gases in lower quantities than coal gasification (8 kg CO_2/kg H_2) [70]. Blue hydrogen, often called low carbon hydrogen, is produced using the steam methane reforming method combined with carbon capture and utilization technology, reducing carbon emissions (1.5 – 5 kg CO_2/kg H_2) [71,72]. Pink hydrogen employs nuclear energy for water electrolysis, red hydrogen utilizes thermal energy from nuclear power for high-temperature catalytic water splitting, also referred to as the thermolysis process, and purple hydrogen is generated through nuclear power plants that facilitate the chemothermal process for water splitting using both nuclear energy and heat [73]. The pyrolysis of methane is now under investigation, primarily at the laboratory scale, yielding solid carbon as a by-product and hydrogen as the principal product, referred to as turquoise hydrogen [74]. This process generates very few greenhouse gas and may be regarded as low-emission hydrogen in the future. Solar energy facilitates water electrolysis for hydrogen generation, referred to as yellow hydrogen [57]. Hydrogen production by water electrolysis or biomass thermochemical processes utilizing renewable resources such as solar, wind, and hydro is referred to as green hydrogen. The benefits of green hydrogen production include the development of bioenergy, enhancement of recycling efforts, achievement of zero carbon emissions, etc [75,76].

Several well-established methods are available for hydrogen production; still, SMR is most common in industries that meet more than 80% of the global hydrogen demand [77]. In the SMR process, the raw material (natural gas) is initially pretreated (removal of sulphur) to reduce the chances of end-product contamination and catalyst deactivation using hydrodesulfurization and activated carbon adsorption techniques. Following pretreatment, steam and methane are reacted at high temperatures (700-1000 °C) and pressure of 3-25 bar [59,78]. The

most common and less expensive catalyst is nickel-based, which produces CO and H_2 (syngas) as the main product with some heat energy as a byproduct. The extra heat generated from the reforming reaction is further used for steam generation. WGS reaction converts further CO and steam to produce more hydrogen, increasing the efficiency of SMR. Pressure swing adsorption is used at the final stage to remove CO_2 and impurities and produce pure hydrogen for further application [79]. The efficiency of the SMR process depends on the hydrogen carbon ratio (H/C) of feedstock and steam to carbon ratio (S/C). Generally, the S/C ratio is 2-3, which means a higher steam supply to increase the efficiency. The typical hydrogen synthesis efficiency in the industrial SMR process is 70-85% [80,81]. The application of SMR-based hydrogen includes transportation fuel, power generation in fuel cells, and feedstock for chemical products such as ammonia, methanol, acetic acid, etc. Despite several applications, SMR remains non-sustainable regarding the endothermic process (higher energy input required), carbon footprint, cost-effectiveness, etc. [82]. Grey hydrogen costs around 1.3 – 2.1 €/kg H_2, and carbon emission is 8-12 kg CO_2/kg H_2 [83,84]. Brown and black hydrogen are comparatively cheaper (1.2-2 €/kg H_2), but larger carbon emission (possibly up to 30 kg CO_2/kg H_2) makes it impractical, looking at the alarming situation of global warming [85,86]. Blue hydrogen somewhere reduces the emission (1.5-5 kg CO_2/kg H_2) but at the cost of expensive (1.5-2.8 €/kg H_2) production [87,88]. The high cost mainly comes from the various purification and carbon capture and storage (CCS) equipment attachments, which lower the overall efficiency by 5-14%, whereas capture efficiency is 53-95% [71,89,90]. Turquoise hydrogen is also a potential candidate with the advantage of an environment-friendly product. It is more expensive (2.55-5 €/kg H_2) than blue hydrogen since electrical energy and feedstock (methane) are more costly than coal [87]. However, solid carbon as a by-product has a market value, making it competitive with other hydrogen production methods [91]. It is more promising than the SMR process, as it is a one step process (pyrolysis) with an energy advantage from 58% less feedstock

needed for the same production [92]. Pink hydrogen production is not common globally, as its energy source (electricity) comes from nuclear power plants, and it is not so common in all places [93].

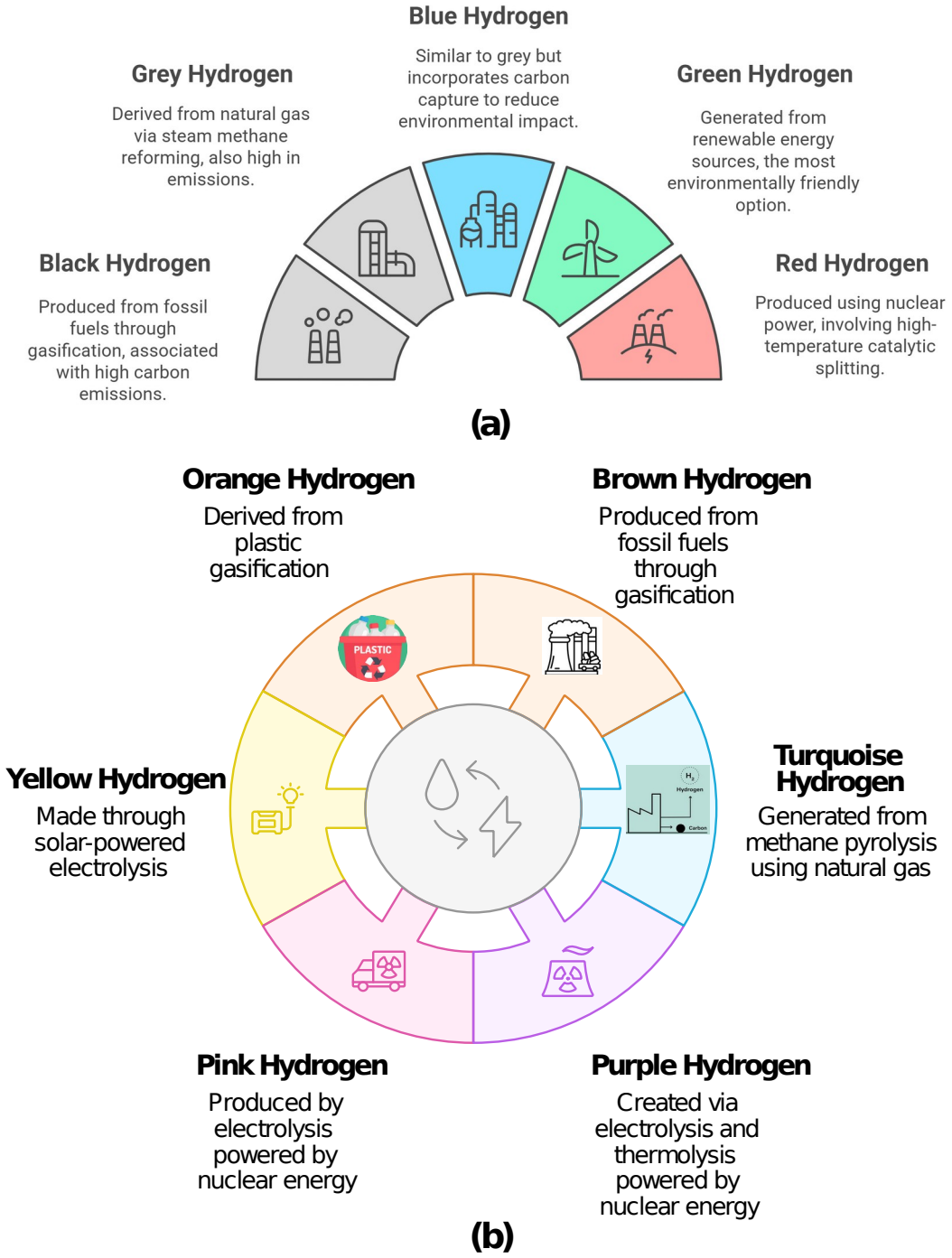
Blue Hydrogen
Similar to grey but incorporates carbon capture to reduce environmental impact.
Grey Hydrogen
Derived from natural gas via steam methane reforming, also high in emissions.
Green Hydrogen
Generated from renewable energy sources, the most environmentally friendly option.
Black Hydrogen
Produced from fossil fuels through gasification, associated with high carbon emissions.
Red Hydrogen
Produced using nuclear power, involving high-temperature catalytic splitting.
(a)
Orange Hydrogen
Derived from plastic gasification
Brown Hydrogen
Produced from fossil fuels through gasification
PLASTIC
Yellow Hydrogen
Made through solar-powered electrolysis
Turquoise Hydrogen
Generated from methane pyrolysis using natural gas
Pink Hydrogen
Produced by electrolysis powered by nuclear energy
Purple Hydrogen
Created via electrolysis and thermolysis powered by nuclear energy
(b)

Figure 5. Classification of hydrogen based on source of raw materials and production methods (a) common type of hydrogen already in practice for mass production (b) type of hydrogen currently under research/pilot scale production.

Green hydrogen is promoted as a future fuel that has the potential to fuel the global energy demand in an eco-friendly manner. Green hydrogen might restore negative climatic externalities; help achieve sustainable development goals and promote circular economy practices to protect natural resources for future generations. Despite numerous cutting-edge advantages, certain limitations remain, like elevated production costs, insufficient dedicated infrastructure, energy loss, and the necessity to assure sustainability, which hinder its commercialization. The current production cost of green hydrogen is 2.5-8.2 €/kg, a major barrier to mass-scale installation for production. It is predicted to reduce costs by 2030 with the maturity of renewable technology and is expected to become competitive with blue hydrogen [58,88,94]. Currently, the primary source of green hydrogen production is electrolysis. There are three well-established and commercialized electrolysis processes that share 70% of the electrolysis market are alkaline water electrolysis (AWE), polymer electrolyte membrane (PEM), solid oxide electrolyser cell (SOEC) with maximum efficiency of 70%, 60%, and 81% respectively [94–96]. Higher purity (>99.95%) hydrogen production is one of the main benefits of electrolysis, but it comes with several drawbacks, such as higher capital cost, higher input energy, instability, delamination of an electrode (a wear process of the layer of material leads to material failure), etc [72,97]. The higher capital cost includes expensive electricity (which holds 90% of the hydrogen production cost), the cost of electrodes, catalysts, membrane, etc. However, there is a prediction showing a cost reduction of electrolysers by 2050 following a reduction in the cost of electricity with the maturity of renewable technology, a hike in the price of natural gas considering carbon emission, and strict rules imposed on the use of only low carbon fuel, etc. The current electrolyzer cost is between 500-1700 US$/kW, and prediction shows below 200 US$/kW [98].

Various novel technologies are under research and development (R&D) to overcome the current problems associated with existing hydrogen production methods. The technologies from anion exchange

membrane, membranelles electrolyzer, photocatalytic generation, dark fermentation followed by photo-fermentation, and plasma arc decomposition are under research for cost reduction and efficiency increment for hydrogen production [99]. Plasma arc decomposition produces hydrogen by passing natural gas through plasma arc and converting it into hydrogen and carbon shoot or from the waste processing [100]. Plasma technologies have proven themselves with time, and their efficiency in working in a wide range of applications is discussed in detail in a further section.

3. Plasma

Plasma, often called the fourth state of matter, is a high-energy and quasi-neutral state characterized by the presence of charged particles, including ions and free electrons [101]. This state is distinct from gases, where atoms or molecules are primarily neutral and interact through short-range forces. In contrast, plasmas exhibit long-range interactions due to the collective behaviour of charged particles, making them highly responsive to electromagnetic fields. The categorization of plasmas into thermal and non-thermal types is based on their temperature and degree of ionization. Thermal plasmas, also known as hot plasmas, are nearly fully ionized and approach local thermodynamic equilibrium (LTE), where the temperatures of electrons (T_e), ions (T_i), and neutral particles (T_n) are approximately equal ($T_e \approx T_i \approx T_n$). These plasmas are commonly produced by arcs, sparks, and flames. On the other hand, non-thermal plasmas have a low degree of ionization, typically less than 1%, and the electron temperature is much higher than that of ions and neutral particles ($T_e >> T_i \approx T_n$) [102]. This non-equilibrium condition allows for applications requiring high selectivity and low thermal damage. For a better understanding of the transition from a gas to a plasma, consider the ionization process. In a gas, atoms or molecules are intact; in a plasma, some electrons are stripped from their atoms, creating ions and free electrons. The plasma state is achieved when the ionization energy is sufficient to overcome the binding energy of electrons in atoms.

Mathematically, the plasma parameter can describe the plasma state, which relates the number of particles within the Debye sphere to the collective behaviour of charged particles. The Debye length (λ_D) is given by,

$$\lambda_D = \sqrt{\frac{\epsilon_0 k_B T_e}{n_e e^2}} \tag{7}$$

where ϵ_0 is the permittivity of free space, k_B is Boltzmann's constant, T_e is the electron temperature, n_e is the electron number density, and e is the magnitude of the elementary charge. This Eq. (7) highlights the importance of electron temperature and density in defining plasma behaviour [102,103].

The unique properties of plasmas make them highly applicable across various fields. Their electrical conductivity and responsiveness to electromagnetic fields are exploited in technologies like plasma TVs and plasma etching. In industrial applications, thermal plasmas are used for metal melting, welding, waste processing, and plasma chemical synthesis due to their high energy density. Non-thermal plasmas, with their low thermal impact, are ideal for surface treatments, sterilization, and pollution control, as they can operate effectively at atmospheric pressure without causing significant thermal damage [104–106]. Additionally, plasmas are used in medical applications for tissue sterilization and food processing to extend shelf life. The ability of plasmas to generate electromagnetic radiation and interact with magnetic fields also makes them crucial in astrophysical studies and controlled fusion research [107]. Overall, the versatility of plasma technology stems from its ability to be tailored to specific conditions, whether thermal or non-thermal, making it a cornerstone in both scientific research and industrial applications.

3.1. Plasma technology for hydrogen production

Non-thermal plasma, despite its numerous advantages, has not yet been integrated into plasma pyrolysis/gasification plants. However, it has been

successfully utilized in reforming various fuels, including diesel, ethanol, methane, and biomass [108–111]. Non-thermal plasma presents a promising approach for reforming and gasifying diverse materials. Nevertheless, its potential for gasifying or reforming municipal solid waste (MSW) in waste-to-energy (WtE) facilities remains unverified. Additionally, integrating a non-thermal plasma gasifier into WtE plant cycles would necessitate modifications to operational conditions and power generation components. Therefore, prior to advancing non-thermal plasma technologies for WtE applications, it is essential to validate the feasibility of non-thermal plasma gasification [112]. In contrast, thermal plasma has been employed for several decades in waste processing and the production of alternative fuels. A more comprehensive discussion on this topic will be provided in subsequent sections.

3.1.1. DC Thermal Arc Plasma

Thermal plasmas, widely utilized in waste treatment, are predominantly generated through direct current (DC) thermal arcs due to their resilience to fluctuations in processing conditions [113]. These arcs are created by establishing a potential difference between two electrodes within a carrier gas environment. The applied electric potential ionizes the gas in the electrode gap, forming an arc that produces thermal plasma. Electrodes are typically constructed from high electrical conductivity materials, such as copper, tungsten, or graphite. Common carrier gases include nitrogen, argon, air, helium, oxygen, and steam [114]. Among these, air is often preferred in waste treatment applications due to its ability to supply adequate oxygen for gasification reactions and cost-effectiveness. However, using oxygen as a carrier gas can exacerbate electrode erosion. Alternative gases like nitrogen, carbon dioxide, and steam are occasionally employed, though they present challenges such as elevated arc voltages and accelerated electrode wear, particularly with steam, which produces a mixture of H_2, O_2, and OH radicals [115]. Electrode erosion remains a significant drawback of DC

arc plasma systems, as the high-temperature regions gradually degrade electrode materials. Although argon can reduce electrode erosion, its lower specific heat and thermal conductivity lead to diminished power output, restricting its utility in waste treatment. Thermal plasma arcs are classified as either transferred or non-transferred as shown in Figure 6, with temperatures typically ranging from 10,000 to 20,000 K. These temperatures are influenced by factors such as input power, carrier gas type, and plasma torch design [116]. Transferred arcs exhibit several advantages over non-transferred arcs, including reduced carrier gas flow rates, higher temperatures (12,000–20,000 K), and adaptability to multi-rod electrode configurations. In contrast, non-transferred arcs generally achieve maximum temperatures of 10,000–14,000 K, with heat dissipation occurring externally to the torch, resulting in lower-temperature interactions with waste materials [114,116].

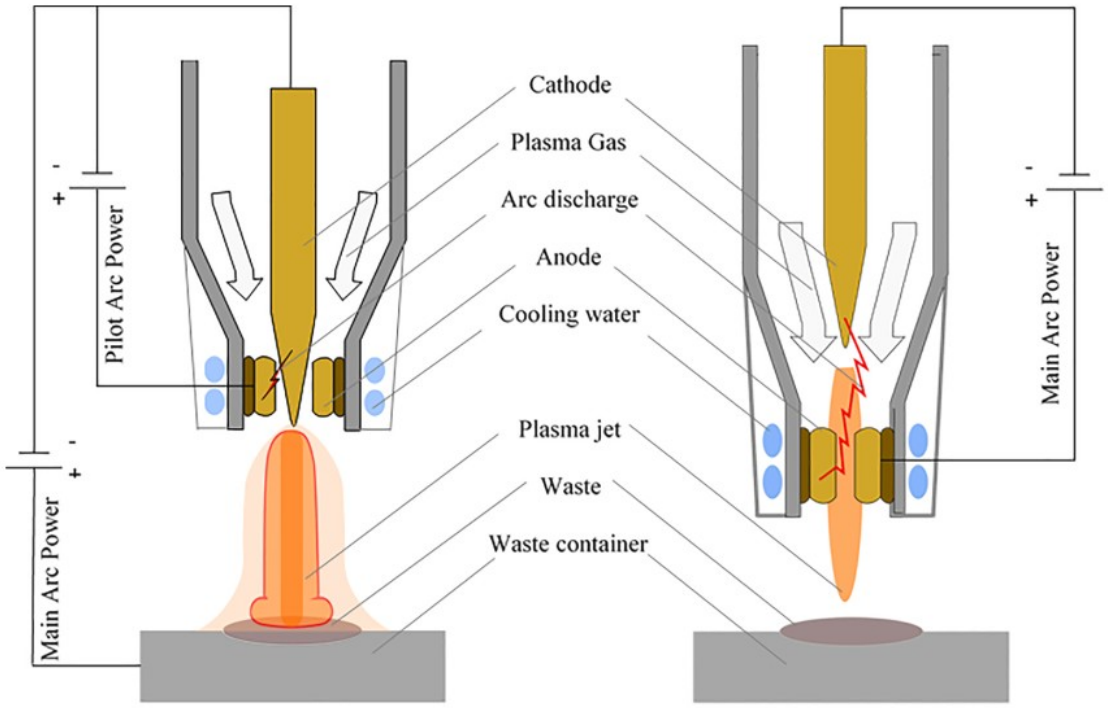

Figure 6. Schematic diagram of DC thermal arc plasma: a) DC transferred thermal arc plasma b) DC non-transferred thermal arc plasma. (Adopted from Ref. ***[114]*** ©Sustainable Energy Technologies and Assessments).

3.1.1.1. DC thermal arc plasma torch for hydrogen production

Various DC plasma torch systems, including Westinghouse, Europlasma, Tetronics, and PSC, are utilized for waste treatment, with operational capacities reaching up to 100 MW [117]. These thermal plasma torches can process diverse waste materials and convert them into valuable products and by-products, as outlined in Table 1. Hrabovsky *et al.* [118] conducted an extensive study on the treatment of waste materials such as plastics, coal, fire sawdust, wooden pellets, refuse-derived fuel (RDF), and pyrolysis oil. The experiments were performed using plasma power ranging from 100 to 140 kW within a chamber volume of 0.2 m^3, maintaining wall temperatures between 1100 and 1400 °C. The highest hydrogen yield (61%) was observed during coal treatment with a steam flow rate of 18 g/min, while the lower heating value of the produced gas was measured at 11.3 MJ/m^3. In another study, Yousef *et al.* [119] employed a DC steam plasma torch to gasify surgical masks (10 kg per run) to generate hydrogen-rich syngas. The process yielded approximately 50% hydrogen and other combustible gases, such as carbon monoxide (22%) and methane (13%), at a power input of 56.9 kW. The syngas production rate was recorded at 3.4 m^3/kg, with a lower heating value of 14.5 MJ/m^3. Ibrahim *et al.* [120] investigated the plasma gasification of municipal solid waste (MSW) within a temperature range of 600 to 1600 °C in a 10 kg/h plasma chamber, using argon as the carrier gas at a flow rate of 10 LPM. The treatment, conducted in the presence of oxygen and steam, favoured carbon monoxide production, with a maximum yield of 95% observed within the temperature range of 1200 to 1400 °C. Tang *et al.* conducted plasma pyrolysis experiments for energy recovery from discarded polypropylene. The nitrogen plasma reactor configures 50 mm in diameter and 1000 mm in height fed with 60 g/min and converted into hydrogen and other combustible gases such as CO, acetylene, methane, etc. The conversion rate of solid waste into gases is 94%, whereas hydrogen is 15% of the composition. The hydrogen fraction is improved by adding steam to 27% with an optimum power supply of 35.2 kW [121]. Similarly, Tang *et al.* [122] investigated plasma pyrolysis of tires to produce hydrogen and carbon black. The tires

fed in the reactor at 80 g/min with a steam supply of 80 ml/min produces 24 % of hydrogen. The steam supply improved the overall gas yield from 42% (without steam) to 77% and hydrogen fraction from 14% (without steam) to 24 % at expense of reduction in solid carbon yield from 58% to 23%. Huang *et al.* [123] investigated plasma pyrolysis of discarded rubber with varies fed rate of 44 g/min to 122.5 gm/min and power input of 30-44 kW. The solid waste rubber is converted into gas (31–77%) and solid carbon (23-69%), where hydrogen percentage is 8-24%. The maximum calorific value of gas is approximately 9 MJ/m^3. Overall, the thermal plasma torch technology for energy recovery in form of higher calorific value gases such as H_2 and CO is found technically and environmentally reliable. The idea of converting waste into a useful resource rather than dumping into landfill or simply converting it into exhaust gases and lower heat value product as incineration and other conventional technologies do is considered as sustainable approach. Looking to the problem of higher power expenses and capital cost the graphite electrode is being in research and practices discussed in detail in next section.

Table 1. Different types of waste treated through DC thermal arc plasma technology

Waste treated	Carrier gas	Power input and feed	H_2 (%)	Reference
Coal	steam	100-140 kW	61	[118]
Surgical mask	steam	56.9 kW, 10 kg/h	50	[119]
Polypropylene	N_2	35.2 kW, 3.6 kg/h	27	[121]
Tire	N_2	35.2 kW, 4.8 kg/h	24	[122]
Rubber	N_2	35.2 kW, 2.6 kg/h	24	[123]
petrochemical	Ar	3.6 kW, 10 cm^3	33	[124]
Carpet	Ar	100 kW, 256 gm	13	[125]

3.1.1.2 Graphite electrode-based plasma pyrolysis/gasification

A variety of materials are employed as electrodes for plasma generation, with copper and tungsten being the most commonly used. Electrodes used in thermal plasma generation must possess high melting points to endure the elevated temperatures generated by the plasma, as lower melting point materials would fail due to melting and evaporation [126]. Tungsten, with a melting point exceeding 3400 °C, is often utilized in thermal plasma torches due to its superior thermal resistance. However, continuous water cooling is essential to prolong the lifespan of tungsten electrodes, resulting in energy losses through waste heat. The water-cooling systems in such torches can consume up to 50% of the total power supply, which could be conserved by employing alternative electrode materials [127]. Graphite electrodes have been explored as an alternative in arc plasma generation, particularly for waste processing applications [25,128]. The non-metal graphite exhibits high electrical conductivity and a melting point above 3000 °C. Its exceptional thermal stability is attributed to its strong covalent bonding, which demands significant energy for bond dissociation. Graphite electrodes are cost-effective, eliminating the need for water cooling and reducing energy requirements compared to tungsten-based plasma torches [25,129,130]. Another advantage of using graphite as an electrode material is that forming carbon monoxide (CO) might enhance the calorific value of the resultant syngas [131]. Unlike tungsten, graphite does not melt under plasma conditions but instead undergoes gradual erosion influenced by factors such as high temperatures environment, oxidation reactions, ion bombardment, and surface morphology. At elevated temperatures, graphite sublimates, converting carbon atoms directly into vapor. Oxidation reactions between carbon atoms and oxygen form carbon dioxide (CO_2), further contributing to material loss. Ion bombardment, caused by collisions of positively charged plasma particles with the graphite (cathode) surface, also leads to erosion. Additionally, surface irregularities and micro-craters can exacerbate uneven erosion. To mitigate these effects, high-density graphite materials are preferred for plasma generation [132,133]. Beyond its susceptibility to erosion,

graphite exhibits advantageous properties such as high thermal conductivity, low thermal expansion, excellent thermal shock resistance, chemical stability, cost-effectiveness, and machinability in various shapes and sizes. These characteristics make graphite a strong candidate for use as an electrode material, potentially overcoming the limitations associated with materials like tungsten [134].

Graphite electrode-based plasma chambers are already being utilized in various waste-to-energy applications. Since plasma torches are costlier than graphite-based electrode torches, developing countries such as India are more interested. Initially, due to higher CAPEX and OPEX, this technology was introduced for processing hazardous waste, such as medical waste. In the last decades, much of the research has been under development on a lab scale to a pilot scale (kg/h), resulting in successful large-scale installations. These all started with the applicability of technology given by Nema and Ganeshprasad for treating medical and plastic waste [135]. Rana *et al*. [25] demonstrated the operation of a pilot-scale plasma pyrolysis plant (horizontal graphite electrodes configuration, 30 kW DC power supply), as shown in Figure 7 with an installed capacity of 50 kg/h, for Municipal Solid Waste (MSW) treatment in Jafrabad, Delhi. The combustible fraction of MSW is processed at approximately 1000 ºC. Gas composition analysis identified carbon monoxide (CO) and hydrogen (H_2) as the primary components, accounting for approximately 30% of the generated gases, with a lower heating value (LHV) of 4.7 MJ/Nm^3. The residues of Ash are characterized using Energy Dispersive X-ray Spectroscopy (EDX) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) techniques, confirming their carbon-free and non-toxic nature. Wastewater from the gas cleaning process exhibited a neutral pH and Total Dissolved Solids (TDS) in the range of 1200–1500 mg/L. These findings underscore the plant's potential as an environmentally sustainable and efficient solution for waste management.

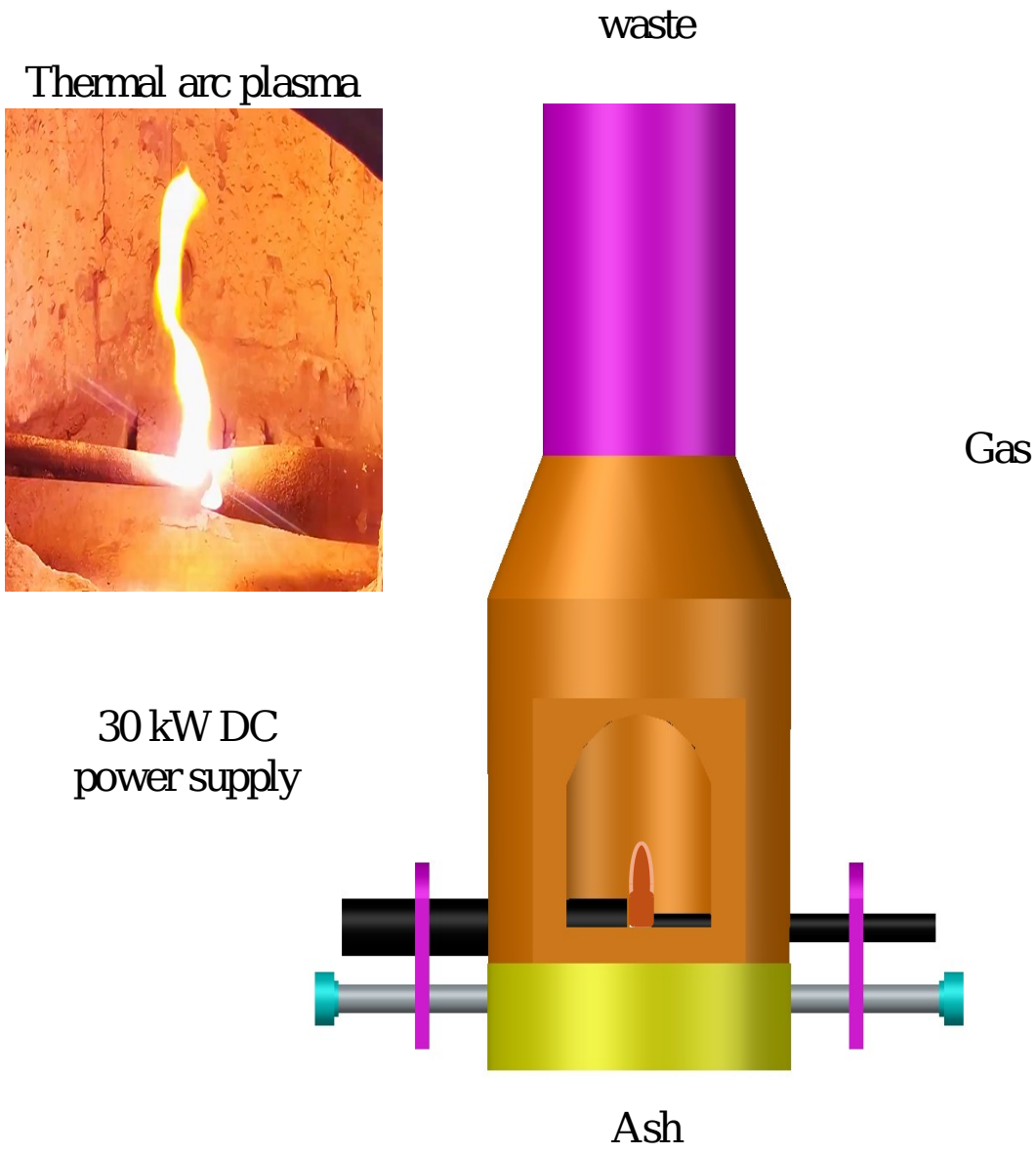

Figure 7. Graphite electrode-based Plasma Pyrolysis plant for MSW processing [25].

Similarly, Yayalik *et al.* [120] treated MSW (10 kg/h) in a plasma gasification chamber (vertical graphite electrodes configuration, 45 kW DC power supply with current setting range of 30-650 A, argon as carrier gas for thermal arc plasma generation with flow rate of 10 L/h), where gas composition study is analyzed for a wide range of temperature between 600-1600 °C with an increment rate of 200 °C measured using Rd-Pt thermocouple (0-1700 °C). The product gas consists of CH_4, C_2H_6, H_2, CO_2, and CO, and their concentration varies with temperature. The plasma gasification experiment conducted for various gasifying agent such as steam (flow rate of 20 L/h, fed at 5 bar of pressure and 100 °C)

and oxygen (flow rate of 20 L/h). The hydrogen concentration found insignificant up to 800 °C, increases from 800-1400 °C mainly for steam as a gasifying agent, and again decreases for further higher temperatures. The reason behind the lower hydrogen concentration is the erosion of the graphite electrode releases free carbon, promoting more hydrocarbon product gases and a low H_2/C ratio of MSW. The suggestion for higher caloric value product gases is to supply more steam at the entrance of higher temperature zones to promote the water gas shift reaction. A more detailed study of the role of graphite electrode erosion during waste treatment is required to find gaps between the reaction mechanism for hydrogen production and how to improve it. Further, Mallick *et al*. [136–138] use a lab-scale graphite-based plasma gasification chamber for processing a variety of waste such as bakelite electrical switches, Acrylonitrile Butadiene Styrene (ABS) plastic, and refuse-derived fuel (RDF). Bakelite switches (40 gm/minute) are converted into syngas (10.2 MJ/m^3) and liquid fuel (31.47 MJ/kg) through CO_2 plasma gasification (CO_2 flow rate of 0.6 LPM), where hydrogen concentration is 24.36% by volume. The maximum syngas yield is 85%, with maximum power consuming 1.6 kW [136]. Similarly, computer keyboard switches made (40 gm/10 minutes) up of ABS plastic are converted into syngas (15.80 MJ/m^3) and liquid fuel (39.13 MJ/kg) using CO_2 plasma gasification (CO_2 flow rate of 0.5 LPM) having volume concentration of hydrogen gas is 30.16% with a total power consumption of 1.12 kW [137]. RDF plasma treatment follows the bakelite and ABS plastic procedure to convert it into syngas (13.95 MJ/m^3) and liquid fuel (30.59 MJ/kg). It achieves a maximum concentration of hydrogen gas (42.6%) with a maximum power consumption of 1.6 kW and cold gas efficiency (CGE) of 49.9% [138]. For all the waste, the common residue component is TiO_2, which has applications in healthcare, paints, and the cement industry. Sharma *et al*. [129] have conducted a detailed simulation study of the thermal performance of a horizontal graphite electrode configuration-based plasma pyrolysis chamber (200 kg/h) using CFD analysis in ANSYS CFX. The study is more about the temperature

distribution inside the chamber, effects of temperature zone on refractory, etc., for the preheating stage and found potential for the waste treatment. As per the discussion of the above-mentioned experiments, the graphite electrode-based plasma chamber has potential for hydrogen production from waste processing. There are still some literature gaps available such as the impact of graphite electrode erosion on product gas composition, interaction of waste with graphite electrode, erosion behaviour with different carrier gases, etc. Since currently there are fewer studies than the literature available for conventional plasma torches, more in-depth studies are required for large-scale installations.

3.1.1.3 Thermal plasma methane pyrolysis for hydrogen production

The limitations of steam methane reforming (SMR) for hydrogen production have prompted the exploration of alternative low-carbon hydrogen production methods. These methods include electrolysis, gasification, pyrolysis, and plasma-based pyrolysis/gasification. However, each approach faces challenges, such as high costs or contamination of the product gas. Historically, industries have employed partial oxidation or thermal decomposition of hydrocarbons like acetylene and ethylene for carbon black production, which is utilized in tire manufacturing as a pigment, UV stabilizer, and conductive or insulating agent in coatings [139]. Several methods are used for carbon black production; details are given in Table 2, including the limitations. Thermal cracking of methane has been identified as a more cost-effective and low-carbon hydrogen production method for carbon black production than partial oxidation, with hydrogen being a secondary product [140]. Methane pyrolysis offers significant advantages, including a reduction of approximately 75% in GHGs emissions (2-3 kg CO_2/kg H_2) compared to the SMR process. Additionally, producing solid carbon black as a valuable byproduct provides an opportunity for supplementary revenue generation, thereby shortening the return on investment (ROI) period for industrial-scale installation [141].

Table 2. Different methods for carbon black production and their limitations [142].

Methods	Product	Limitations
Thermal decomposition	Carbon black	• H_2 produced is used for reactor heating rather than as an energy source for other applications. • Cost is competitive with SMR; thus, commercialization is challenging.
Thermo-catalytic decomposition	Carbon black, H_2	• Fat deactivation of metal catalysts, Lifetime of catalysts found in order of few hours.
Molten metal bath decomposition	Carbon black, H_2	• Low-quality carbon black as it is difficult to separate and recover from the molten metal.
Concentration solar decomposition	Carbon black, H_2	• Product yield is poor from indirect heating • Solar concentrators are costly • Quality control of carbon black is challenging
Non-thermal plasma decomposition	Carbon black, H_2	• Research is at a very early stage • Successful commercialization is not achieved

The thermal decomposition of methane is an endothermic reaction, as given in Eq (8) , necessitates temperatures exceeding 900 °C [143,144]. The use of solid catalysts can lower this temperature requirement. However, during the process, carbon particles may accumulate on the catalyst's surface and diffuse through it, leading to the gradual deactivation of active sites. Consequently, catalyst deactivation adversely impacts the hydrogen production rate, and the recovery or regeneration of the catalyst involves significant costs [145,146]. Methane dissociates at 500 °C, with complete dissociation occurring at 1000 °C. Equilibrium data indicate that at temperatures between 1000 °C and 2500 °C, hydrogen and carbon black are the predominant products of methane decomposition [139]. A key challenge in methane pyrolysis for hydrogen production lies in achieving the high temperatures required for complete

conversion. At these temperatures, the formation of unsaturated hydrocarbons such as acetylene, ethylene, and benzene occurs more rapidly than their complete decomposition into hydrogen and solid carbon. The nucleation mechanism for soot formation involves the generation of polycyclic aromatic hydrocarbons (PAHs) [147]. Adding unsaturated hydrocarbons like acetylene to PAHs increases molecular weight and leads to hydrogen deficiency through hydrogen abstraction, resulting in primary soot particle formation. These primary particles grow further with acetylene decomposition. However, preventing soot nucleation remains challenging, particularly under rapid cooling conditions. A simplified equilibrium diagram for one mol of methane, excluding solid carbon as a species, suggests the theoretical formation of 98.5% acetylene at 1875 °C [139]. Exploiting this theoretical possibility, electrical arc discharge or plasma processes have been developed for acetylene production. Later, several researchers demonstrated that plasma-driven methane pyrolysis can effectively produce carbon black and hydrogen from methane [148–151]. Plasma methane pyrolysis can produce hydrogen at high conversion efficiency, low cost, and lower emission than the SMR process. SMR process converts natural gas into hydrogen with a conversion efficiency of only 50% (0.5 kg H_2/kg CH_4) in addition to CO_2 emission of 9-12 kg/kg H_2 production [152].

$$CH_4 \rightarrow C\ (s) + 2H_2 \qquad \Delta H° = 74.91\ kJ/mol \tag{8}$$

Research on non-thermal plasma pyrolysis remains in its early stages, with various technologies, such as “gliding arc” and “cold arc” plasma, being proposed. Unlike non-thermal plasmas, thermal plasmas achieve ionization rates sufficient to induce Joule heating, making them highly adaptable and tunable heat sources. These plasmas are particularly suited for endothermic processes and can operate without direct CO_2 emissions [142]. The commercialization of technology dates back over a century, beginning with Rose's 1920 patent [153]. Several carbon black manufacturers built upon this original patent between 1920-1990. In the

1990s, the Norwegian engineering company Kvaerner (now Akker-Kvaerner) developed DC plasma technology (tested 3 MW plasma pilot plant in 1992) for the co-production of carbon black and hydrogen from natural gas. Kvaerner constructed an industrial plant in Karbomont, Canada, in 1997 with a capacity of 20,000 tons of carbon black and 6,000 tons of hydrogen per year. However, the project was discontinued in 2003 due to technological challenges and poor quality of the carbon black byproduct [154,155]. Fulcheri *et al.* [156] introduced a novel concept of plasma methane pyrolysis for CO_2-free co-production of carbon black and hydrogen in 1995. These advancements have since been the subject of numerous patents and scientific studies. Monolith Materials, Inc., a California-based start-up, undertook industrial development of plasma-driven methane pyrolysis in collaboration with the PERSEE team at MINES-ParisTech between 2012-2018. A pilot scale facility is successfully developed and tested in 2018. The facility has a production capacity of 14,000 tons of carbon black and 4,600 tons of hydrogen per year, as shown in Figure 8 [142].

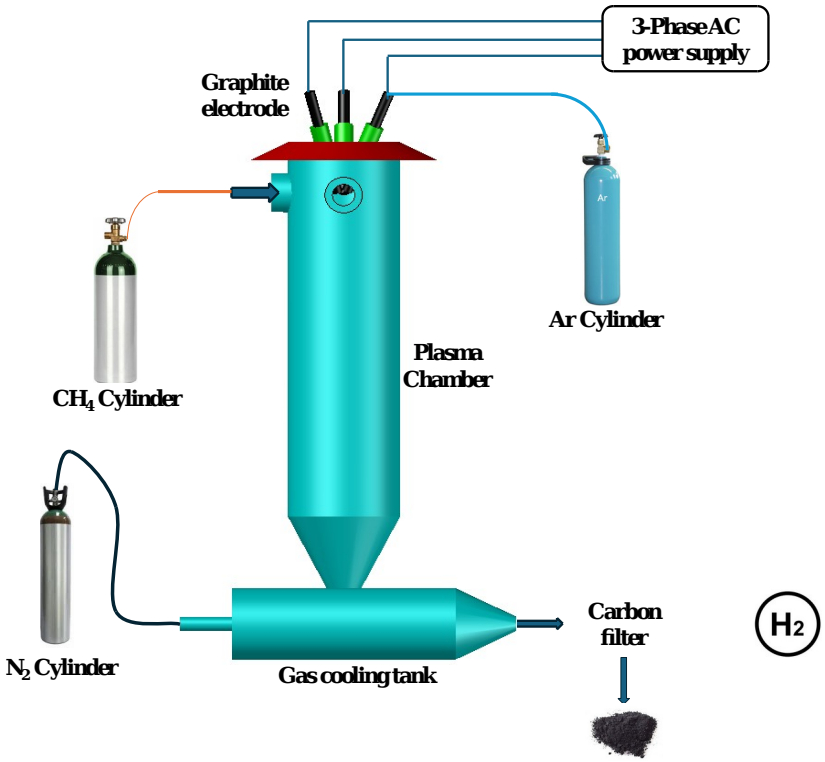

Figure 8. Plasma methane pyrolysis technology for turquoise hydrogen and carbon black production [142].

3.1.2 AC thermal arc plasma for hydrogen production

Parallel to the DC thermal plasma, AC plasma is also in practice. In this category, a combination of electrode systems uses different AC phases with frequency majorly in the range 50-60 Hz. The torch consists of cylindrical geometry with electrodes at an oblique angle to the gas flow. The plasma gas passes through the annular space between the electrodes, where electric power input is dissipated via the Joule effect, generating a high-temperature plasma plume. This energy is transferred to the main flow through convection, diffusion, and radiation. The resulting plasma plume typically exhibits extremely high velocities (several hundred meters per second), driven by the narrow nozzle design and the intense thermal expansion of the heated plasma gas. In this system, multiple arcs coexist within a large discharge chamber, creating a less confined plasma than classical DC plasma torches. This configuration is sustained by a continuous, high concentration of charge carriers due to the relatively long lifetime of ionized species despite alternating current and zero-current crossings. The alternating change of electrodes between the anode and cathode reduces erosion, thereby improving performance resource factors and thermal efficiency [157]. Consequently, the plasma achieves a lower mean temperature and operates in a diffuse mode to some extent, unlike the contracted mode in DC systems. Arc motion, driven by alternating currents and magnetohydrodynamic (MHD) forces, enhances convective heat transfer. External magnetic fields can control arc behaviour, such as spot motion to reduce electrode erosion and arc elongation to regulate voltage and power. Energy is distributed through gas heating, relaxation of excitation, and atom recombination, reducing radiation and convection losses. These features result in higher electro-thermal efficiency compared to DC plasma torches.

The earlier attempt at AC arc plasma research started during the mid-20th century in the United States and the former Soviet Union, followed by Norway, France, Germany, and Japan [158,159]. Intending to simulate

a high-temperature environment during the re-entry of ballistic missiles in a laboratory, then aeronautical engineers and scientists worked around the arc heater (plasma arc jet) to achieve high gas enthalpy flows. For the initial decade, foundational work on the modelling AC arc column was performed by Philips [160] and Geister [161]. They laid down initial design criteria for AC arc heaters at high-pressure operations (>10 atm) and provided critical insights for device development. Several joint collaboration research programs were initiated, such as the decarbonization of cement powder and the development of a 500kW electro burner to explore its applicability to an industrial scale, but were halted due to economic reasons [162]. Many researchers like Fabry and Rehmat studied the influence of gas flow characteristics on arc motion. They found that with the presence of three electrodes, mutual electromagnetic coupling between arcs extensively modifies the flow in reactors. Rehmat [163,164] continued exploring the possibilities under different parametric studies such as current frequency and gas flow rate on the arc behaviour, through theoretical and experimental approaches. T Watanabe *et al*. [165] developed multiphase AC plasma torches for glass melting applications. With twelve tungsten electrodes arranged 30° apart with a diameter of 3.2 mm powered by 40-50 kW and a current drawn of 290 A, multiple arcs can co-exist simultaneously in the electrode gap region.

Pioneer research in the development of AC torches is carried forward by the Rutberg group of the Institute for Electro-physics and Electric Power at the Russian Academy of Sciences in St Petersburg (IEE RAS). They introduced two types of stationary multiphase AC plasma generators designed for waste destruction and syngas production. Their study detailed plasma generators operating in oxidizing media, with average power levels of up to 50 kW and high-power systems reaching 500 kW. Figure 1 The plasma electron density n_e recorded were in the range of $10^{17}\,cm^{-3}$. The power supply provided a high current-low voltage and a high-voltage, low current stage for average and high-power mode,

respectively, along with reactive power compensation. Using air as an oxidizing media in the range of 2-25 g/s for average power and 10-70 g/s for average power settings, a thermal efficiency of approximately 90 % with an electrode lifetime of 100 hr. They worked on a multiphase AC plasma generator that moderates heat content at the outlet 1.5 to 12.5 MJ/kg. The average temperature recorded with airflow can be controlled from 1500-6000 K [159]. Another work by Rutberg *et al.* [166] presented models of high-voltage plasma torches in a long cylindrical channel using 10 kV open circuit voltage. Typical parameters include a current drawn of 40-85 A; a voltage drop of 2.5-3.2 kV with a flow of 60-100 g/s yields an arc length exceeding 2 m. Geometrical sizes of arcs depend on the electrodes' placement, starting from the minimal gap between electrodes and the wall casing, blown away through gas flows to expand in the ambient and provide stability.

Regarding waste reduction and processing, Rutberg *et al.* [167] use a mixture of H_2O/air in 1:6 at a constant mass flow for plastic waste gasification. The process parameter follows a voltage drop of 1.0-1.8 kV current of 28.5A, with power of 52-86 kW, resulting in an average arc length of 798 mm. Using a thermodynamic equilibrium model, estimated arc temperatures were 10,000-11,500 K and arc diameter of approximately 4.47 mm. The experiments reveal a reduction in arc temperature and electrical conductivity with a rise in steam content in the gas flow. Energy consumed in the plasma gasification is 11 - 12.3 MJ/kg, which results in a syngas yield of 3.62 – 3.48 m^3/kg, the composition of H_2 is in range of 55.5 - 62.5 by volume percent and CO is 32.8 - 34.1 volume percent for a range of steam content of 54.20 - 85.29 by mass percent. Surov *et al.* [168] use a mixed gas composition of steam, air, CO_2, and CH_4 as a working gas to decompose wood, coal, and refuse-derived fuel (RDF). A combination of one or two plasma torches up to 50 kW is used. The temperature of the product gas and ash does not exceed 1200℃. The percentage of H_2 and CO obtained is in the range of 43-55 by volume percent. The maximum arc length measured is 0.5 m. A snapshot of the device in operation is shown in Figure 9. Messerle *et al.*

[169] use an electric arc plasmatron for medico-biological wastes. Applied power across the device varies from 11 to 22.5 kW with plasma forming gas feed of 0.8 to 3.8 g/s and of current strength 170A to 260A. The mass of charged waste is from 5 to 7 kg for a duration of 30 min. The effluent gas comprised 63.4 % CO, 6.2 % H_2, 29.6 % N_2, and 0.8 % S by volume. The total synthesis gas (CO & H_2) concentration was 69.6 volume percent. The main gasification agent is air, followed by saturated water vapour as a secondary agent. With steam content of almost 50%, the synthesis gas obtained has proportions of CO ~ 35% and H_2 around 42% by volume in the exhaust at a temperature of 1300 K. Kumkova *et al.* [170] used a three-phase AC plasma torch for the processing of Carbon tetrachloride (CCl_4) and chlorobenzene (C_6H_5Cl) with steam and CO_2 as plasma-forming gas. Under high-temperature steam, methane undergoes reforming, producing hydrogen and carbon monoxide. Power used in the range from 106 – 116.6 kW with high voltage up to 10 kV. Upon decomposition of organic matter, the hydrogen binds with the chlorine present in it. The composition of products includes H_2, CO_2, CO, HCl, and CH_4, where H_2 in range of 32.44-45.05 % and CO in range of 12.97 – 45.51%. The product gas temperature varies from 1250-1400 K, effective for methane steam reforming. Li *et al*. [171] observed a similar product, which showed that for pyrolysis of chemical waste (mixture of different classes of chlorine bi phenyls), the main gaseous products are C, CH_4, H_2, and HCl with a rise in temperature up to 1000 K, a significant percentage increase in H_2 followed by a decrease in C, CH_4, and HCl at the exhaust/outlet. Major wastes processed using thermal plasma powered by AC source are listed in Table 3.

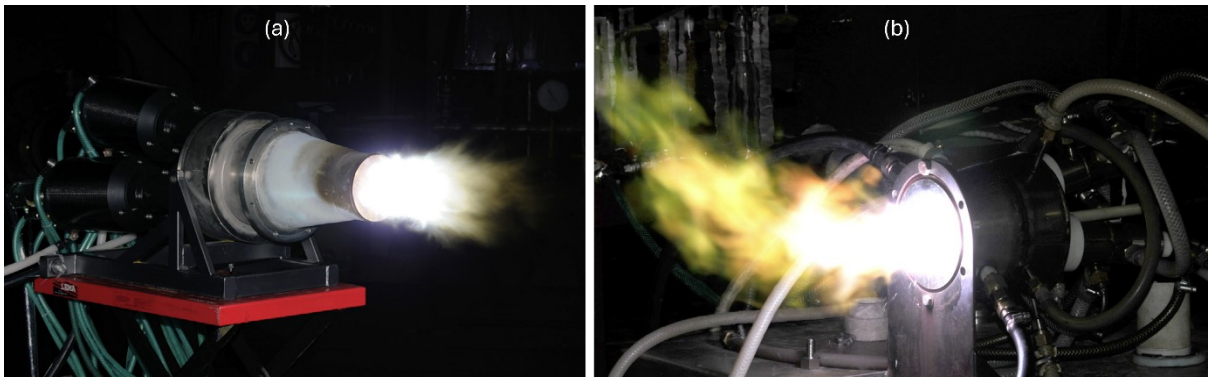

Figure 9. a) High voltage 410 kW AC plasma torch b) multi-gas 3 phase arc AC plasma torch at 100W. (Adopted from Ref. [168] ©Elsevier).

Table 3. Different wastes treated through AC thermal plasma.

Waste treated	Carrier gas	Power input and feed	H_2 (%)	References
Plastic waste	Steam, air	57.65–87.54 kW	62	[167]
Coal	Steam, CO_2, CH_4	5-150 kW	22.96	[172]
Wood	Steam, CO_2, CH_4	5-150 kW	28.20	[172]
RDF	Steam, CO_2, CH_4	5-150 kW	16.95	[172]
Medico-biological waste	Air	35kW, 5-7 kg	42	[169]
Polystyrene	Steam, Air	350 kW, 110 kg/h	< 50	[173]
MSW/Tires Mix	O_2	3.22 kW/kg	22.62	[173]
Wood residues	Air, CO_2, O_2, steam	0.37-2.88 kW/kg	11.1-54.6	[174]
Organo-halogen substances	Steam and CO_2	106-116.6 kW	32-44	[170]

3.1.3 RF-ICP thermal plasma

Radio Frequency Inductively Coupled Plasma (RF-ICP) systems generate plasma by utilizing high-frequency alternating current, typically within the range of 2 to 27 MHz [175]. This current flows through a coil

surrounding the plasma chamber, inducing an electric field that, in turn, initiates a circular or ring-shaped current within the reactor, as shown in Figure 10. This process leads to plasma formation, characterized by exceptionally high energy densities ranging from 10^5 to 10^7 W/cm^2 and operating temperatures spanning from approximately 1000 K to 10,000 K. The extreme thermal environment and substantial ionized particle concentrations of 10^{16} to 10^{17} particles/cm^3 facilitate rapid reaction kinetics by promoting interactions among free ions, excited species, and neutral particles. These conditions significantly enhance heat and mass transfer processes, making RF-ICP systems highly effective for waste pyrolysis and gasification applications, where efficient energy transfer and high reaction rates are essential [176,177].

A major advantage of RF-ICP technology is its ability to operate with a diverse range of working gases. These include conventional noble gases like argon and helium, and reactive gases such as oxygen, hydrogen, nitrogen, and even hydrocarbon-based gases like methane. Among these, argon remains the most widely used due to its favourable ionization properties at high temperatures, relatively high atomic mass, and cost-effectiveness. Additionally, its density contributes to superior thermal conductivity, enabling efficient energy transfer within the plasma [101]. However, maintaining gas purity is crucial, as even trace impurities can lead to plasma instability, torch quenching, or operational failure. To optimize performance and overcome the density-related limitations of argon, lighter gases such as helium, hydrogen, or nitrogen are often introduced into the system. These gases enhance the axial velocity of the plasma jet, improving the system's overall efficiency and enabling a more focused, directed plasma output [178,179]. Another significant advantage of RF-ICP technology is its scalability to high power levels exceeding 1 MW, making it suitable for large-scale industrial applications requiring continuous operation. Unlike DC plasma systems, which often suffer from electrode erosion, current-carrying limitations, and excessive cooling requirements, RF-ICP systems operate contactless, eliminating these constraints. This design ensures longer operational lifespans, greater

stability, and higher throughput, making RF-ICP a preferred choice for high-temperature processing applications across various industries [180].

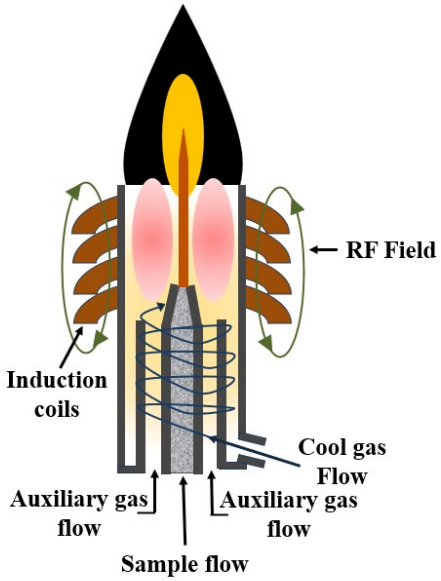

Figure 10. Schematic diagram of RF-ICP thermal plasma [175].

RF-ICP technology has emerged as a highly effective method for processing waste materials, including heavy oil, tire-derived powder, and MSW blended with raw wood. One of the key advantages of RF-ICP is its flexibility in utilizing different carrier gases, with nitrogen (N_2) being widely employed due to its stability and ability to facilitate controlled plasma reactions. The power input required for these waste treatment processes spans a broad range, from as low as 0.3 kW to as high as 10 kW, allowing customization based on the nature and composition of the waste. Recent studies have demonstrated the capability of RF-ICP systems to decompose complex waste materials and optimize the generation of valuable gaseous products, particularly hydrogen (H_2), which is of growing interest as a clean energy carrier. Tang *et al.* [181] investigated the pyrolysis of waste tires operating at power levels between 1600 and 2000 W under controlled pressures ranging from 3000 to 8000 Pa. Their experimental setup successfully generated a reactive plasma environment with temperatures varying between 1200 K and 1800 K. By employing a dual-electrode system, the study enhanced the yield of hydrogen (H_2) and carbon monoxide (CO), key components of

syngas, demonstrating the potential of RF-ICP plasma for hydrogen-rich gas production. Similarly, Aboughaly *et al*. [182] developed an advanced RF-ICP-based thermal plasma pyrolysis system that achieved an impressive 89 wt.% conversion efficiency without the formation of tar, a common issue in conventional pyrolysis methods. Their system incorporated an 8-turn copper RF-ICP torch within a 12 L thermochemical reactor, ensuring stable operation for over 30 minutes at peak plasma temperatures reaching 2000 °C. Pyrolysis experiments were conducted at temperatures between 550°C and 990°C for extended durations, while gasification processes operated at approximately 1300°C with a residence time of just 1 second, leading to high hydrogen production efficiency. Further advancements in RF-ICP technology have been demonstrated by Shie *et al*. [177], who explored the gasification of municipal solid waste blended with raw wood. Their findings revealed syngas yields ranging from 88.59% to 91.84 vol%, with energy recovery efficiencies varying between 59.07% and 111.89%, highlighting the capability of RF-ICP plasma to maximize hydrogen output while minimizing energy losses. Tang *et al*. [183] demonstrated the effectiveness of an RF capacitively coupled plasma reactor for plastic gasification, operating within a pressure range of 1000–10,000 Pa and temperatures of 1500–2000 K. Their study revealed that nitrogen plasma yielded up to 12 vol.% hydrogen, while steam plasma significantly enhanced hydrogen production, with the combined H_2 and CO content reaching 28 vol.%, highlighting the system's potential for syngas generation. Similarly, Tu *et al*. [184] investigated RF plasma pyrolysis of rice straw, emphasizing its ability to enhance gas production while minimizing tar formation. The study reported a substantial improvement in hydrogen yield at elevated temperatures, reinforcing RF plasma technology's efficiency in producing hydrogen-rich syngas. Further supporting the role of plasma-assisted hydrogen production, Nishikawa *et al*. [185] explored the gasification of carbonaceous waste using a hybrid thermal plasma system with steam. Their findings demonstrated that the water-gas shift reaction facilitated significant weight reduction

of the feedstock while enhancing hydrogen production. The results showed that steam-assisted plasma treatment generated more hydrogen than other thermal plasma techniques, reinforcing its potential for efficient waste-to-hydrogen conversion. Building upon these advancements, RF-ICP thermal plasma has shown immense potential for enhancing hydrogen (H_2) production efficiency by leveraging its unique ability to generate highly reactive species at relatively lower bulk gas temperatures. Unlike conventional thermal plasma processes, which rely primarily on high temperatures for waste decomposition, RF-ICP thermal plasma facilitates targeted molecular dissociation and selective gas-phase reactions through energetic electrons and excited species. This mechanism significantly enhances hydrogen yield while minimizing undesirable byproducts such as tars and solid residues. Error: Reference source not found summarizes the various waste treated with RF-ICP with outcome as hydrogen gas.

Despite their many advantages, RF-ICP systems also come with certain limitations. One of the key challenges lies in their energy transfer efficiency, which is inherently lower than that of Direct Current (DC) plasma systems. This inefficiency primarily stems from the limitations of high-frequency power sources, as conventional RF-ICP setups often utilize vacuum tube-based Class C-tuned oscillators with efficiencies below 50%. However, recent advancements in solid-state power supplies, capable of operating at frequencies approaching 450 kHz, have significantly enhanced the performance and energy efficiency of high-power RF-ICP systems. Optimizing parameters such as waste injection points and gas flow dynamics is critical for ensuring stable plasma operation and maximizing system efficiency [116]. Hybrid plasma systems have been developed to address individual plasma configurations' limitations, leveraging the strengths of both RF and DC plasma technologies [186]. Keun Su Kim *et al.* [187] introduced an innovative hybrid DC-RF thermal plasma process to produce high-quality carbon black and pure hydrogen. This system leverages the combined advantages of both plasma types by utilizing high-velocity plasma jets

generated by a DC torch, which are then expanded and further energized within an inductively coupled RF torch.

Table 4. Different types of waste treated through RF- ICP technology.

Waste treated	Carrier gas	Power input and feed	H_2 (%)	Reference
Tire	N_2	1.8 kW, 0.012 kg/h	47	[181]
Rice Straw	N_2	0.3-0.7 kW	2.2 mg/L	[184]
Tire Powder	N_2	1.6-2 kW, 0.012 kg/h	-	[188]
MSW and wood	N_2	10 kW, 0.2 kg/hr	88.59 - 91.84 vol %	[189]
Plastic (PE)	N_2, Steam	2 kW, 0.4 g/min	12 vol.%	[183]
Charcoal	Ar, Steam	34 kW,	280 - 3300 ppm	[185]

3.1.4 Microwave thermal plasma

Among various Atmospheric Pressure Plasma Jets (APPJs), Microwave Atmospheric Pressure Plasma Jets (MW-APPJs) stand out for operating at significantly higher frequencies up to the GHz range (300 MHz-10 GHz). These are the high-frequency electromagnetic waves commonly employed in applications like hydrogen production, waste treatment, CO_2 dissociation, etc. Extensive research has explored continuous and pulsed power modes to generate these MW-APPJs with different geometries, including waveguide- and non-waveguide-based, as Rath et al. discussed [190]. MW heating is preferable for application purposes because of the high ionization rate with mostly electrodeless operations. This electrode-free configuration (waveguide-based geometries) guarantees an extended operational lifespan and minimizes the need for maintenance. Microwave (MW) plasmas demonstrate substantially higher electron densities than low-frequency plasmas. The elevated electron density facilitates extensive dissociation of the working gas, resulting in a highly energetic and reactive environment. These advantages make MW-APPJs suitable for applications like hydrogen production, and they have gained significant attention due to their production effectiveness and increasing efficiencies. Several literatures discuss hydrogen production with MW-

APPJs. A schematic diagram of the microwave plasma setup as shown in Figure 11.

A large-scale gasifier used for hydrogen-rich synthetic gas production from Indonesian brown coal using microwave-generated steam plasma, as investigated by Uhm *et al*. [191]. The process, operating at temperatures up to 1700°C, demonstrated nearly complete carbon conversion (almost 100%) and achieved a hydrogen-rich synthetic gas efficiency of 84%, for total production of 500 kW of syngas. The compact and efficient design of the system highlights its potential for integration into medium-scale power generation facilities. Another investigation by Khongkrapan *et al*. [192] shows microwave plasma pyrolysis of refuse-derived fuel that generates combustible gas containing approximately 14% hydrogen (H_2). The product gas also contains about 66% carbon monoxide (CO) and 4% methane (CH_4). This process presents a potentially marketable form of clean energy. These findings indicate that plasma-assisted pyrolysis is a viable waste-to-energy conversion technology, producing marketable clean energy products. Yoon *et al*. [193] studied Glycerol gasification using microwave plasma torches to produce syngas with high hydrogen content. High carbon conversion and cold gas efficiency can be achieved by maintaining the O_2/fuel ratio below 0.4. Increasing microwave power enhances hydrogen and CO content, improving syngas heating value, carbon conversion, and cold gas efficiency. Supplying glycerol through a gas-liquid nozzle increases hydrogen and CO content while decreasing CO_2. Lin *et al*. [194] investigates hydrogen production through microwave plasma pyrolysis of Spirulina algae. Experiments were conducted at varying microwave power levels, yielding hydrogen gas in the 36.75-45.13% (volume fraction) range. The selection of power levels significantly impacts hydrogen conversion, with 20.62-52.43% of hydrogen atom mass content in dry algae being converted to hydrogen gas. These findings enhance the understanding of Spirulina algae pyrolysis and hydrogen yield, informing the development of hydrogen production technologies. Hydrogen (H_2) production via microwave-assisted gasification of sewage

sludge is investigated as a promising alternative to traditional methods, addressing concerns regarding non-renewable resource consumption and greenhouse gas emissions by Hu *et al.* [195]. The study systematically examines the influence of gasification agents, specifically steam and CO_2, on syngas production and the generation pathways of H_2. The research highlights that a steam-CO_2 mixed atmosphere enhances syngas yield and quality. The maximum H_2 concentration of 63.73 vol% was achieved at a steam/CO_2 ratio 36, demonstrating a synergistic effect between the two agents. A study numerically simulates a downdraft plasma gasifier to analyze syngas production with plasma reactions showing impacts of the equivalence ratio on syngas properties, which were investigated by Ibrahimoglu *et al.* [196]. The syngas mainly contains H_2, CO, and CH_4 and can be used for direct combustion, and the findings reveal that increasing the equivalence ratio from 0.20 to 0.45 reduces the syngas's lower heating value. Delikonstantis *et al.* [197] explore air/N_2 gasification of industrial fermenter byproducts in a microwave plasma reactor for organic compound valorization. The research identified an operating window for stable operation, achieving up to 89% carbon conversion efficiency and 41% cold gas efficiency. The syngas product composition was H_2:CO:CO_2 in proportion of 41:53:6. Hong *et al.* [198] explores syngas production via brown coal gasification using a 2.45 GHz microwave torch plasma. The study highlights the plasma's ability to enhance chemical reactions, eliminating the need for catalysts. Experiments involving Indonesian brown coal with a high ash content yielded a gas composition of 48% hydrogen, 23% carbon monoxide, 25% carbon dioxide, and 4% methane at a coal-to-steam ratio 1.36. These findings suggest the potential of microwave plasma technology for efficient gasification of low-grade coal, offering a compact alternative to traditional methods. Production of hydrogen using different waste treatment with microwave plasma are summarized in Table 5.

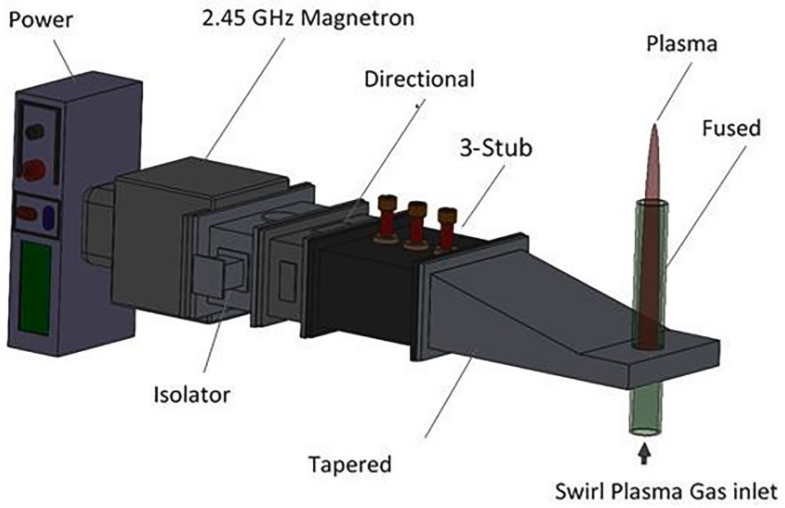

Figure 11. Schematic diagram of Microwave thermal plasma. (Adopted from Ref. *[114]* ©Elsevier).

Table 5. Hydrogen production through Microwave plasma.

Waste	Carrier gas	Power input and feed	H_2 (%)	References
Polyethylene	Ar	0.8 kW, 5 g/run	9.5	[192]
RDF	Ar	0.8 kW, 5 g/run	13.8	[192]
Glycerol	N_2	1-2 kW, 180 g/h	57	[193]
Spirulina algae	N_2	0.8-1 kW	22.8	[194]
Sewage sludge	CO_2	1.4 kW, 10 g/run	31.74 – 35.28	[195]
Coal	Air	10 kW, 100 kg/h	33.6	[196]
Biomass	Air/N_2	6 kW	41	[197]
Brown Coal	Steam	4 kW	48	[198]

4. Discussion

Plasma technology, due to its robustness, fast reaction, and high energy densities of thermal plasma sources for waste decomposition, is considered a clean, efficient and environmentally friendly method with potential alternatives to traditional incineration, landfills, and composting. However, this technology is in its early stages and needs significant efforts to streamline a standard set of waste volume reduction procedures and energy recovery at the exhaust. Detailed techno-economic feasibility for the pilot plant is one more challenge to look for. In this section, all these challenges are discussed in terms of technical,

economic, and policy scope, and a solution is also proposed to solve the current problem.

4.1. Technical & economic scope

Plasma gasification is an advanced waste-to-energy technology that converts waste into valuable products such as syngas, hydrogen, and vitrified slag. Despite its potential, several challenges hinder its widespread adoption. Figure 12 outlines key challenges and their possible solutions, as explored in the following sections

1) *High Capital and Operational Costs*

Plasma gasification requires significant initial investment due to the high cost of plasma torches, reactors, and auxiliary systems. Operational costs are also high because of the energy-intensive nature of plasma generation and maintenance. This makes it less economically viable compared to conventional waste management methods. Addressing the challenge, integrating plasma gasification with renewable energy sources such as solar or wind to reduce operational costs and improve sustainability is suggested. Additionally, scaling up and adopting modular designs can reduce capital costs, making the technology more accessible for small-scale applications. Government subsidies and financial incentives, as highlighted by Heberlein & Murphy (2008), might also encourage investment in plasma gasification projects [116].

2) *Feedstock Variability and Pre-Treatment Requirements*

The heterogeneous nature of waste, including municipal solid waste (MSW), industrial waste, and biomass, poses challenges in maintaining consistent gasification performance. Variability in moisture content, particle size, and composition can affect the efficiency of the process. Pre-treatment of feedstock, such as drying, shredding, and sorting, is a proposed solution that could improve homogeneity and process efficiency [199]. Adaptive plasma systems that dynamically adjust to varying feedstock properties using real-time monitoring and control systems have also been proposed [200]. Co-

gasification is another solution that involves mixing different types of waste (e.g., biomass with plastic), which could stabilize the process and improve syngas quality [201].

3) *Syngas Quality and Contaminant Removal*

The syngas produced during plasma gasification may contain contaminants such as tar, particulate matter, and harmful gases (e.g., HCl, H2S), which can damage downstream equipment and reduce the usability of the gas. Advanced gas cleaning systems, including cyclones, scrubbers, and filters, can effectively remove contaminants [117]. Catalytic tar reforming, which uses catalysts to break down tar into simpler gases, has been shown to improve syngas quality [202]. High-temperature filtration systems, such as ceramic filters, can also remove particulate matter efficiently.

4) *Energy Efficiency and Process Optimization*

Plasma gasification is energy-intensive, and achieving high energy efficiency remains challenging. The energy required to generate plasma can sometimes outweigh the energy recovered from the process. Optimizing plasma torch design using alternative gases (e.g., argon, nitrogen) and optimizing power input can improve efficiency [203]. Heat recovery systems, such as heat exchangers, can recover waste heat from the process and use it for preheating feedstock or generating electricity [174]. Process integration, such as combining plasma gasification with other waste treatment technologies (e.g., anaerobic digestion), can maximize energy recovery.

5) *Environmental Concerns and Emissions*

Although plasma gasification is cleaner than traditional incineration, still the emissions of toxic gases such as NOx, dioxins, and heavy metals is debatable, raising environmental concerns. Thus, including advanced flue gas treatment systems, such as selective catalytic reduction (SCR) and activated carbon injection, can reduce emissions [204]. Vitrification of residues, a byproduct of plasma

gasification, can produce inert slag that can be used in construction materials, reducing landfill requirements [200]. Life cycle assessment (LCA) studies can also help identify and mitigate environmental impacts throughout the process.

6) *Lack of Commercial-Scale Demonstrations:*

Most plasma gasification projects are at the pilot or laboratory scale, and there is a lack of large-scale commercial demonstrations, which limits confidence in the technology. Public-private partnerships (PPPs) are one of the solutions that can encourage collaboration between governments, industries, and research institutions to fund and develop commercial-scale projects [116]. Similarly, documenting successful pilot projects and sharing best practices can build confidence and attract investors [199]. Regulatory support, such as clear guidelines and standards for plasma gasification, can facilitate its adoption [117].

7) *Public Perception and Acceptance*

Public resistance to waste treatment technologies, often due to a lack of understanding or fear of emissions, can hinder the deployment of plasma gasification plants. Community engagement and education campaigns can help the public understand the benefits of plasma gasification, such as reduced landfill use and lower emissions. Transparent reporting of environmental and operational data can build trust and demonstrate the safety and efficacy of the technology [201]. Hosting open days and tours of pilot plants can showcase the technology in action [200].

Plasma gasification holds immense potential for waste-to-value processing, particularly in the context of the growing hydrogen economy. However, addressing the challenges of high costs, feedstock variability, syngas quality, energy efficiency, emissions, and public perception is critical for its successful implementation. Research and innovation, policy support, and public engagement can pave the way for plasma gasification to become a cornerstone of sustainable waste management and clean energy production.

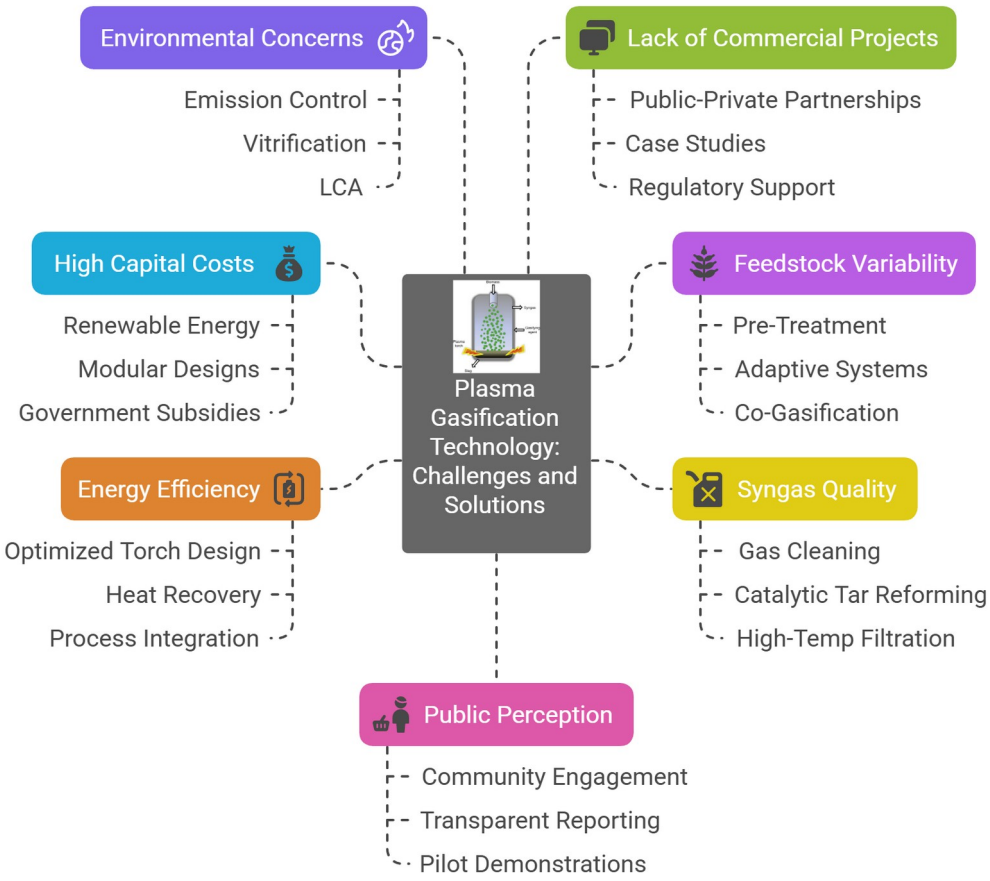

Figure 12. Challenges associated with the commercialization of plasma pyrolysis/gasification technology and solutions proposed for better outcomes.

4.2. Policy Scope

India's waste management framework has evolved significantly over the decades, reflecting the growing need for sustainable practices. Figure 13 shows the evolution of rules and laws underlined by government of India for handling and processing of wastes. The journey began with the Water (Prevention and Control of Pollution) Act 1974, which, although primarily focused on water pollution, indirectly addressed waste management issues [205]. Followed by the Environment (Protection) Act 1986, which provided a broader framework for managing hazardous waste and other environmental concerns [206]. The Hazardous Waste (Management and Handling) Rules, 1989, further emphasized the safe storage, treatment, and disposal of hazardous waste. However, the first significant milestone in municipal solid waste management came with the Municipal Solid Waste (Management and

Handling) Rules, 2000, which introduced structured guidelines for the segregation, collection, transportation, processing, and disposal of municipal solid waste (MSW). These rules emphasized waste reduction, recycling, and composting and mandated local authorities to set up waste processing and disposal facilities [207]. In 2016, the Solid Waste Management Rules replaced the 2000 guidelines, marking a significant shift towards more comprehensive waste management practices. These rules introduced advanced concepts like extended producer responsibility (EPR), waste-to-energy, and decentralized waste management. They also mandated source segregation of waste into biodegradable, non-biodegradable, and hazardous categories and encouraged public-private partnerships (PPPs) for waste management [208]. Parallelly, in 2011 the Plastic Waste (Management and Handling) Rules, were introduced to address the growing plastic waste problem, focusing on recycling and proper disposal. These were revised in 2016 to include a ban on certain types of plastics and mandate thicker plastic bags for better recyclability while emphasizing EPR for plastic producers. Similarly, the E-Waste (Management and Handling) Rules, 2011, regulated the disposal and recycling of electronic waste and were expanded in 2016 to include producers, consumers, and recyclers, with specific collection targets for producers [209]. The Construction and Demolition Waste Management Rules, 2016, provided guidelines for the recycling and reuse of construction waste, further strengthening India’s waste management framework. The most recent update came in 2022 with the Updated Solid Waste Management Rules, which further emphasized waste-to-energy technologies, including incineration, gasification, and pyrolysis, and encouraged the use of advanced technologies for waste processing, aiming to achieve zero-landfill targets [210].

Despite these advancements, the commercialization of plasma gasification technology, a cutting-edge waste-to-energy solution, remains limited in India due to high capital costs, regulatory barriers, and lack of awareness. To address these challenges, specific modifications to the

existing guidelines are essential. Firstly, plasma gasification should be explicitly included in waste-to-energy policies, with clear implementation guidelines and emission standards. This would provide a regulatory framework for its adoption to emphasize the need for policy support to promote advanced waste-to-energy technologies. Secondly, financial incentives such as subsidies, tax breaks, and low-interest loans should be offered to reduce the high capital costs and attract private sector participation. Zhang *et al.* [202] underscore the role of financial incentives in scaling up advanced waste-to-energy technologies. Thirdly, the regulatory approval process for plasma gasification plants should be streamlined, and a single-window clearance system should be established to reduce bureaucratic hurdles. Mountouris *et al.* discuss the importance of regulatory support for waste-to-energy projects, emphasizing the need for a simplified approval process. Additionally, research and development (R&D) support is crucial for addressing technical challenges and improving the efficiency of plasma gasification. Funding should be allocated for R&D to optimize the technology and reduce operational costs, as suggested by Sanlisoy & Carpinlioglu, who highlight the role of co-gasification and feedstock pre-treatment in improving process efficiency [201]. Furthermore, specific emission standards for plasma gasification plants should be developed, and continuous monitoring should be mandated to ensure environmental compliance and build public trust. Gomez *et al.* [200] emphasize the importance of emission control and monitoring in plasma gasification, noting that clear standards are essential for environmental sustainability. Lastly, public awareness campaigns should be launched to educate the public about the benefits of plasma gasification and address misconceptions. Community engagement is critical for successfully implementing new technologies, highlighting the role of public acceptance in promoting waste-to-energy projects.

By integrating these modifications, India can harness the potential of plasma gasification to achieve its waste-to-energy and sustainability goals. The technology aligns with India’s objectives of reducing landfill dependency and promoting clean energy production. With policy support,

financial incentives, streamlined regulations, R&D funding, emission standards, and public awareness, plasma gasification can become a cornerstone of India's waste management framework, paving the way for a cleaner and healthier society. Since few plants are successfully running across the globe, a framework should be laid down to improve performance. Government policies can boost the adaption of these technologies through the obligation to industries to offset a certain portion of their dumped waste using thermal plasma.

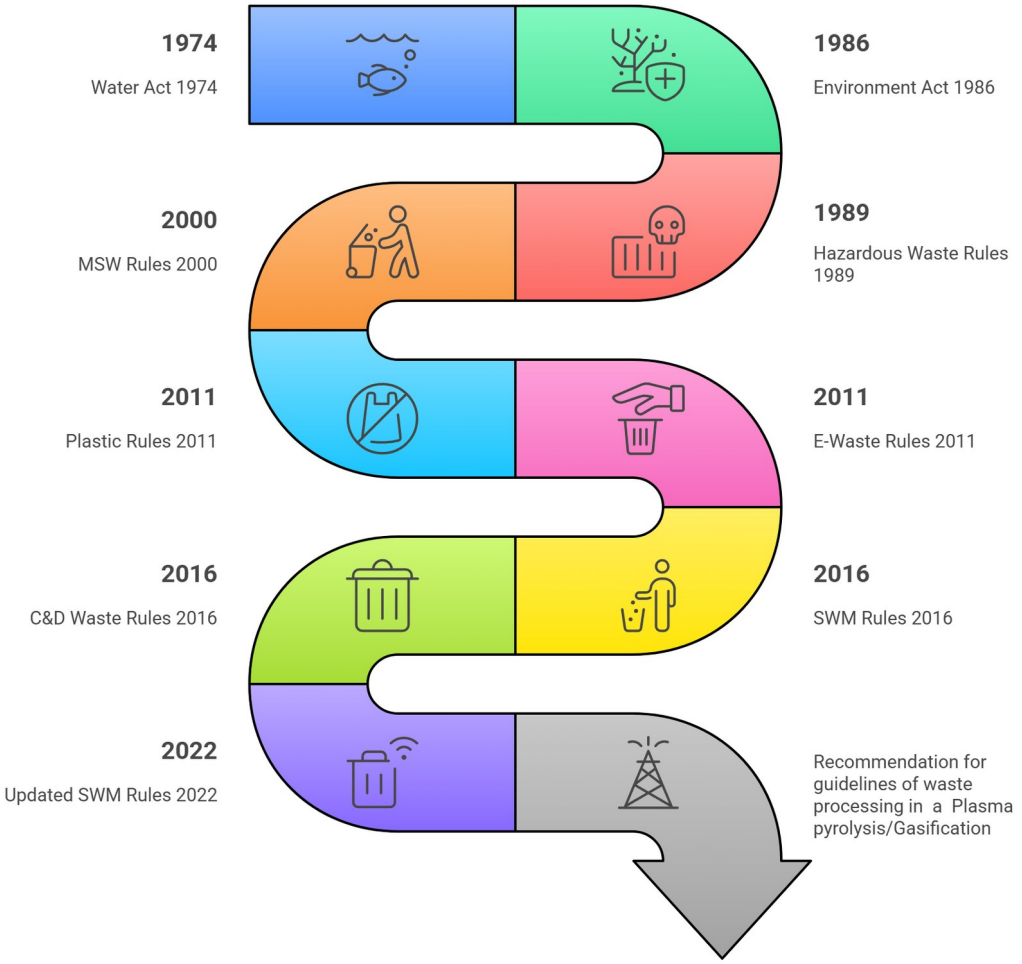

Figure 13. Evolution of waste management rules and regulation by Indian government for handling and processing of waste with year leading to modifications required for plasma gasification technology commercialization.

4.3. Net zero concept exploiting plasma gasification technology: a sustainable solution

Over the past decade, plasma gasification technology has emerged as a clean and environmentally friendly solution for waste management,

offering a sustainable pathway to address the growing global waste crisis. The concept of achieving net zero waste, which converts waste completely into valuable resources without generating secondary waste, can be realized by integrating plasma gasification with other advanced waste treatment technologies. Figure 14 depicts a systematic process for realization of net zero concept. The process begins with waste generation and segregation, where waste is categorized into biodegradable (e.g., food waste, agricultural residues) and non-biodegradable (e.g., plastics, textiles, and other combustible materials) fractions. Biodegradable waste to biogas plants, where anaerobic digestion converts organic matter into biogas, primarily composed of methane (CH_4) and a nutrient-rich residue. The residue has application as compost for agricultural purposes. It reduces the volume of waste sent to landfills and produces renewable energy in the form of biogas. The non-biodegradable and combustible fraction of waste can then be processed using plasma gasification technology, which thermally decomposes waste at extremely high temperatures (up to 5,000°C) into syngas (a mixture of hydrogen and carbon monoxide), vitrified slag, and heat. The syngas produced gas utilization in multiple ways. For example, it can be directly combusted in syngas generators to produce electricity, converted into hydrogen through the water-gas shift reaction, or used as a raw material to produce chemicals such as methanol and ammonia [37]. The vitrified slag, a glass-like byproduct, is inert and can be used in construction materials, ensuring that no secondary waste is generated [200].

Further enhancing the efficiency and sustainability of this integrated system, the biogas generated from anaerobic digestion can be utilized in plasma methane pyrolysis. The process that decomposes methane into hydrogen and solid carbon without emitting CO_2. It is also known as turquoise hydrogen production, boosting the hydrogen economy and providing a carbon-neutral pathway for hydrogen generation [201]. Integrating biogas plants with plasma gasification creates a synergistic effect, where the methane from biogas is used to produce hydrogen, and the residual heat from plasma gasification can be harnessed to optimize

the anaerobic digestion process, thereby improving overall energy efficiency. A hybrid approach combining anaerobic digestion, plasma gasification, and plasma methane pyrolysis offers a comprehensive solution for achieving net zero waste. By converting waste into high-calorific-value gases (syngas and hydrogen), chemicals, and construction materials, this system ensures that no waste is left untreated or sent to landfills. The concept of net zero is further reinforced by the complete utilization of residues, such as vitrified slag, which can replace traditional construction materials, reducing the demand for natural resources and minimizing environmental impact [117]. Additionally, hydrogen production through plasma methane pyrolysis and syngas conversion aligns with global efforts to transition to a hydrogen-based economy, providing a clean and renewable energy source for various industrial and transportation applications [202].

Integrating these technologies addresses the challenges of waste management and contributes to carbon neutrality by reducing greenhouse gas emissions. For instance, biogas in plasma methane pyrolysis avoids CO_2 emissions associated with traditional hydrogen production methods, such as steam methane reforming. Moreover, the energy recovered from syngas and biogas can offset the energy requirements of the waste treatment process, making the system self-sustaining and economically viable [174]. In conclusion, integrating anaerobic digestion, plasma gasification, and plasma methane pyrolysis offers a transformative approach to achieving net zero waste. The hybrid system ensures complete waste utilization without generating secondary waste by converting waste into valuable resources such as hydrogen, electricity, chemicals, and construction materials. Adopting such advanced and integrated technologies not only addresses the global waste crisis but also supports the transition to a sustainable and circular economy, paving the way for a cleaner and greener future.

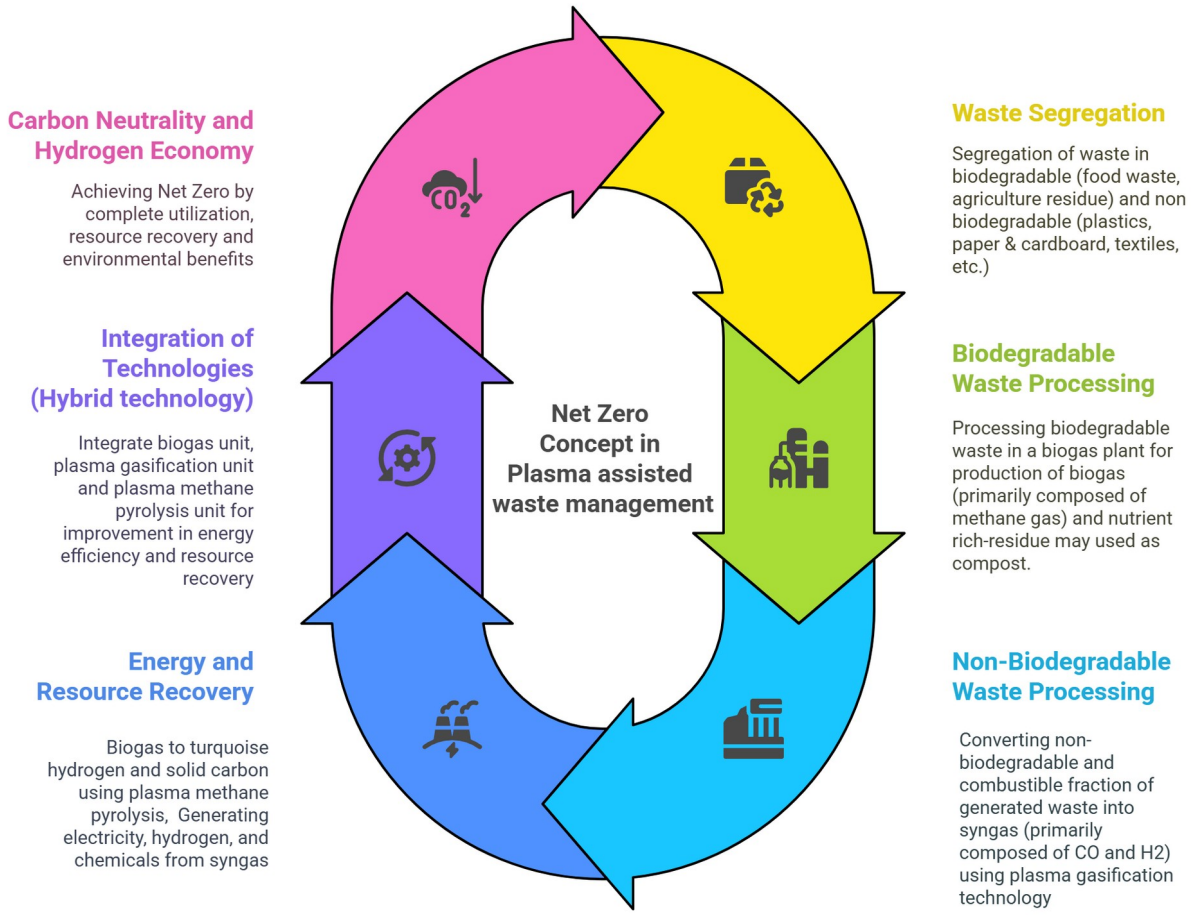
Carbon Neutrality and Hydrogen Economy
Achieving Net Zero by complete utilization, resource recovery and environmental benefits
Integration of Technologies (Hybrid technology)
Integrate biogas unit, plasma gasification unit and plasma methane pyrolysis unit for improvement in energy efficiency and resource recovery
Energy and Resource Recovery
Biogas to turquoise hydrogen and solid carbon using plasma methane pyrolysis, Generating electricity, hydrogen, and chemicals from syngas
Net Zero Concept in Plasma assisted waste management
Waste Segregation
Segregation of waste in biodegradable (food waste, agriculture residue) and non biodegradable (plastics, paper & cardboard, textiles, etc.)
Biodegradable Waste Processing
Processing biodegradable waste in a biogas plant for production of biogas (primarily composed of methane gas) and nutrient rich-residue may used as compost.
Non-Biodegradable Waste Processing
Converting non-biodegradable and combustible fraction of generated waste into syngas (primarily composed of CO and H2) using plasma gasification technology

Figure 14. Net zero concept exploiting plasma pyrolysis/gasification technology for complete utilization of generated with better efficiency and resource recovery leading to carbon neutral environment.

5. Conclusion

Waste was never the problem our perception of it was. For too long, it has been seen as the end of the line, when in fact, it holds the potential to be the beginning of something meaningful. The review covers diverse aspects related to MSW waste processing with current technologies. It emphasizes the prospects of plasma technologies for catering to complex loads with the possibilities of sustainable energy recovery. Operating factors influencing hydrogen production yield through different plasma processes is presented. Further, it discusses the technical and economic scope with policy recommendations for adapting plasma technologies in managing MSW. Significant insights from this comprehensive assessment of the literature are outlined below:

- The generation of MSW, its buildup in landfills, and the resulting environmental impacts will continue to be a critical issue unless significant changes are made.
- Advancing traditional waste management practices to plasma-based technologies represents a sustainable strategy for waste valorization and clean energy production.
- Unlike conventional approaches, plasma technology, particularly thermal plasma, efficiently converts various waste streams into valuable products like hydrogen/syngas, supporting low-carbon solutions aligned with global sustainability objectives.
- Plasma technology offers a progressive alternative to traditional landfilling and gasification methods, with its capability to generate clean hydrogen from waste positioning it as a pivotal component in advancing sustainable industrial practices.
- Various plasma sources offer distinct advantages depending on the waste type, and optimizing operational parameters can significantly enhance process efficiency and hydrogen yield.
- Successful commercialization hinges on addressing three key factors:

  (a) Technological scalability, which involves optimizing plasma systems to handle diverse waste types without compromising performance

  (2) Economic viability, achieved by minimizing operational costs through energy-efficient designs and capitalizing on by-products such as hydrogen and recovered materials; and

  (3) Supportive policy frameworks, including subsidies, carbon credits, and regulatory incentives to promote widespread adoption.
- Integrating multiple technology for net zero concept: combining conventional methods like anaerobic digestion with advanced processes such as plasma gasification and plasma methane pyrolysis enables the treatment of mixed waste streams at a single location, resulting in more environmentally sustainable and economically valuable products.

- Through strategic investments, enabling policy frameworks, and ongoing technological advancements, plasma-assisted waste-to-energy systems have the potential for large-scale deployment, offering both environmental and economic advantages in the pursuit of sustainability.

In light of growing energy demands and the persistent challenge of municipal solid waste, this review highlights plasma-based waste-to-hydrogen technologies as a sustainable alternative aligned with circular economy principles. By enabling clean and efficient energy recovery, these methods support the development of a greener and healthier society. Optimistically, this review will inspire new perspectives on waste management to ensure environmental well-being and a better quality of life for all living beings, which aligns with the global Sustainable Development Goals.

Authors' contributions

Tejashwi Rana: Conceptualization, Methodology, Investigation, Formal Analysis, Writing – original draft**. Aishik Basu Mallick:** Conceptualization, Methodology, Validation, Formal analysis, Writing – original draft. **Radhika T P:** Conceptualization, Investigation, Visualization, Formal Analysis, Writing – original draft. **Suryasunil Rath:** Conceptualization, Investigation, Formal analysis, Writing – original draft**. Pratyay Asim Chattopadhyay:** Investigation, Writing – original draft. **Satyananda Kar:** Supervision, Validation, Writing – review and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data Availability Statement

Data will be made available upon reasonable request.

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